

## **NANOMAGIQC**

IST-2001-33186

Nanotechnology and Magnetic Qubits to Implement Quantum Computation

# **Final Report**

Covering period 01.01.2002-31.12.2004

Report Preparation Date: 15.12.2004

Classification:

Contract Start Date: 01.01.2002 Duration: 36 months

Project Co-ordinator: UB Partners: NPL, CSIC, HPLB



Project funded by the European Community under the "Information Society Technologies" Programme (1998-2002)

# Table of Contents

1.	Executive summary	3
	1.1. The consortium	
	1.2. Summary of objectives and achievements	6
2.	Project objectives	13
	2.1. Year 1	13
	2.2. Year 2	13
	2.3. Year 3	14
	2.4. Management and co-ordination objectives	15
3.	Methodologies	16
4.	Project results and achievements	23
<i>5</i> .	Deliverables and references	31
	<i>3.1. Deliverables.</i>	31
	3.2. References	
6.	Potential impact of project results	45
<i>7</i> .	Future outlook	49
8.	Appendix 6. Project's achievements fiche	52

## 1. Executive summary

#### 1.1. The consortium

#### a. Partners

<u>The Consortium</u> consisted of four contractors from two countries representing major research institutions involved in material preparation, quantum phenomena and communication engineering, each partner playing an indispensable role in the project:

- Universitat de Barcelona, Spain (UB). Partner leader and consortium co-ordinator: Prof. Javier Tejada.
- National Physical Laboratory, Teddington, UK (NPL). Partner Leader: Dr. Patrick Josephs-Franks.
- Consejo Superior de Investigaciones Científicas, Spain (CSIC). Partner Leader: Prof. Fernando Briones.
- Hewlett-Packard European Laboratories, Bristol, UK (HPLB). Partner leader: Dr. Tim Spiller.

The collaboration consisted of both experimentalists (UB, NPL, CSIC) and theoreticians (HPLB):

- In the UB group there are scientists formed in the preparation of magnetic particles and clusters and physicists with large experience in the detection of low temperature quantum phenomena.
- The NPL group has more than twenty years of experimental experience in applications of SQUIDs and the Josephson effects in areas of precise measurement applications.
- The CSIC group comprised two units:
  - The first unit, Instituto de Microelectrónica de Madrid, (IMM), has accumulated for years a significant expertise in the deposition of thin films (oxides and superconductors), electron beam and optical lithography, reactive ion beam etching, AFM manipulation and local oxidation, processing and characterization of magnetic dots.
  - The second unit, Institut de Ciència de Materials de Barcelona (ICMAB), has expertise in the field of molecular magnetism, specifically in the synthesis and characterization of single-molecule magnets based on molecular clusters.
- The HPLB group is expert in quantum computation and information and during the last years was working on new quantum magnetic concepts.

#### b. Roles of the partners

The roles of the partners in the Consortium were briefly the following:

#### • <u>UB</u>: Workpackages 2, 3, 4, 6, 8, 9.

Scientists belonging to the Magnetics Laboratory were responsible for preparing ferri- and antiferromagnetic nanoparticles and magnetic clusters. The magnetic characterisation of materials and the study of feasible measurement set-ups for the microwave and rf resonance experiments allowing the detection and measurement of the magnetic qubits was also the contribution of this group.

Most of the microwave experiments were performed in Barcelona in collaboration with Dr. Joan Manel Hernandez who was contracted as postdoc by the group of CSIC in Madrid. The expertise of Dr Hernandez in the field of microwave, waveguides and cavities was crucial to the work performed in Barcelona.

Finally, being responsible for the co-ordination of the project, UB undertook the management and administration and dissemination and implementation duties.

#### • NPL: Workpackages 1, 5, 6.

The Fundamental & Wavelength Standards team was involved in research into precision measurement techniques involving, amongst others, superconducting electronics (SQUIDs, Josephson oscillators), cryogenic UHV STM, AFM and precision magnetic resonance (electronic and nuclear spins). This team was responsible mainly for SQUID development and evaluation and was also involved predominantly in nanoparticle and molecule positioning developments.

The NPL group was working very hard in the characterization of small SQUIDs to be attached to the  $Mn_{12}$  samples in order to better study both the variation of the magnetization occurring prior to the emission of radiation and the frequency spectra of the emitted radiation.

#### • <u>CSIC</u>: Workpackages 1, 2, 3, 5, 6.

The design and obtaining of molecular clusters as well as the preparation of thin films with them was carried out by the Molecular Magnetic Materials Laboratory of the ICMAB unit, under the supervision of Prof. Jaume Veciana. This unit has been working very hard to get new molecular magnets to test the influence of the decoherence phenomena in the suppression of the radiation emission.

The development of nano-techniques and technologies necessary for the fabrication, handling, positioning and integration of a single magnetic qubit into a superconducting circuit element containing a microSQUID were carried out within the IMM unit. The approach focussed on the extension of existing expertise in the physics and fabrication of nanostructures (based on epitaxial semiconductors and metals and the manipulation by AFM techniques) into the field of superconducting and magnetic nanostructures.

It is also important to mention the enormous consulting work done by Prof. Fernando Briones (CSIC-IMM) for solving many different experimental problems the group of Barcelona had to face to arrive to the detection of radiation.

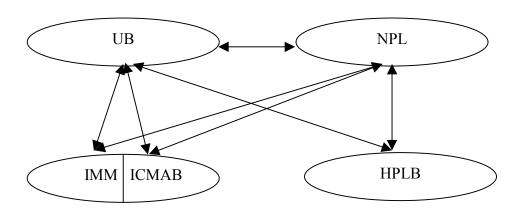
#### • <u>HPLB</u>: Workpackage 7.

The theoretical part of the collaboration was lead by the HPLB group. This group has many years of experience in both QIPC theory and quantum phenomena in condensed matter systems.

#### c. Collaborations

#### Summary of inner collaborations:

- UB-NPL: microwave measurements, superconducting ring, SQUID measurement results.
- NPL-CSIC-ICMAB: nanomagnet materials, SPM measurements, exchange of materials, results and personnel.
- CSIC-IMM-NPL: micro-SQUIDs and micro-Hall devices, testing exchange of results and personnel.
- CSIC-IMM-UB: micro-SQUIDs and micro-Hall probes, exchange of devices, results and personnel.
- UB-CSIC-ICMAB: nanomagnets, exchange of results and samples and personnel.
- HPLB–NPL–UB: SQUID and nanomagnet characteristics for modelling input, and modelling of nanomagnet-microSQUID dynamics.
- UB–NPL–HPLB: Proposals for experiments to give useful physical parameters.
- Exchange of personnel between all partners.



#### Summary of external collaborations:

- NPL-Wernsdorfer: micro-SQUIDs exchange of devices, feedback to modify devices.
- NPL-Chalmers University: exchange of personnel, micro-SQUID device, testing of SQUIDs, e-beam lithography devices.
- NPL-Lam CSIRO: micro-SQUID devices, exchange of devices and results.
- NPL—Cambridge University: state-of-the-art e-beam lithography devices (< 4 nm).
- HPLB–University of Cambridge: system + detector + environment modelling.
- HPLB–University of Bristol: weak measurement modelling.
- CSIC-ICMAB-CNR Bologna: Nanopatterning of SMMs on different surfaces.

## 1.2. Summary of objectives and achievements

The two most important objectives of this project as originally stated in the "Description of work" document (Annex 1 to contract) were the search of the so-called single molecular magnets (SMMs) and their implementation in a SQUID circuit for testing their possible operation as magnetic qubits.

On the basis of the reviewers report of the results obtained along the first year, the original project goals turned out to be too ambitious for a three-year project and were subsequently redefined to focus on the exploration of the fundamentals of quantum nanomagnets and the development of measurement tools for them.

Workpackages 1, 5 and 6 were therefore largely revised and two of them were even renamed to be more realistic: WP5 changed from "Realisation of logic gates" to "Development of measurement tools for quantum nanomagnets", and WP6 changed from "Qubit measurement" to "Exploration of the fundamentals of quantum nanomagnets" (see the "Revised working plan for years 2 and 3" (June 2003) and the appendix to the "Periodic Progress Report No. 2" (January 2004)).

According to this, the work developed during years two and three focussed mainly on the detection and characterization of the microwave radiation emitted from molecular magnets and the preparation and characterization of microSQUIDs.

A detailed list of the specific project objectives can be found in section 2, while a yearly breakdown of the achievements of the project follows next.

#### a. Year 1

The work performed during the first twelve months of this project developed as planned in the "Description of work" document (Annex 1 to contract) with minor deviations that were conveniently addressed.

Several microSQUIDS and micro-Hall probes were designed by the NPL group and the CSIC- IMM unit. The designs were such as to allow to demonstrate the sensitivity of the SQUID to their loop size. Initial difficulties found by CSIC-IMM with the e-beam processing of sputtered Nb films were solved and complete devices with loop area below  $1\,\mu\text{m}^2$  were fabricated and transferred to UB and NPL for characterization. Epitaxial InAs/GaSb shallow channel Hall microprobes were fabricated by MBE, e-beam lithography and RIE, and attempts to precise positioning magnetic dots on the cross area of the probe were successful. A second set of SQUID designs were carried out by NPL to futher improve their characteristics based on the first set of results.

Even smaller SQUIDs were also obtained from two other sources, that use different fabrication techniques but should allow for loops sizes to be produced that are beyond those using optical lithography techniques. An in-house ability was also set up at NPL to bond the SQUIDs to various sample holders and to enable to pattern our own microSQUIDs using electron-beam lithography and local oxidation using an atomic force microscope. All this work was complemented by the CSIC-IMM unit with the

fabrication of magnetic dots accurately positioned off center of the micro-Hall probes by optical lithography.

The CSIC-ICMAB unit prepared and characterized several new  $Mn_{12}$  Single-Molecule Magnets (SMMs) that overall differ on the nature and size of the organic ligands located at the inner coordination sphere of the cluster. Such ligands, which range from openshell radicals to chiral ligands, were carefully chosen both on the quest for synergistic effects and/or the development of new characterization techniques complementary to magnetic detectors. This work, which was successfully achieved, was developed in close collaboration with the UB group and the CSIC-IMM unit. In a second step, two new methodologies to place individual SMMs on a microSQUID area were developed. The first one was the preparation of nanocomposite thin films, made from a polycarbonate polymeric matrix and  $Mn_{12}$  molecules, followed by treatment with different organic solvent vapours. In addition, nanolithographic techniques (wetting/dewetting) were used to prepare arrays of  $Mn_{12}$  SMM on a Si/SiO<sub>2</sub> surface. Finally, preliminary experiments on controlled crystallization processes using supercritical conditions were performed in order to produce  $Mn_{12}$  crystalline clusters.

The systems synthesised by the CSIC-ICMAB unit were characterised by the UB group, which also explored the use of microwave measurements to further investigate and characterise molecular magnets. In this sense, the UB group worked to show that crystals of molecular nanomagnets exhibit enhanced magnetic relaxation when placed inside a resonant cavity. Strong dependence of the magnetization curve on the geometry of the cavity was observed, providing evidence of the coherent microwave radiation by the crystals. The spin state of molecular clusters was also investigated using microwave cavities and resonators under photon absorption conditions. The tunneling probability depends on both the temperature and magnetic field and photons may be absorbed by the spin levels in the two potential wells of the anisotropy when the systems are inside cavities. The competition between the spin-phonon interaction and the interaction between the spin system with the electromagnetic ac field associated to the photons was studied.

The two CSIC units (ICMAB and IMM) collaborated in order to synthesize Single-Molecule Magnets (SMMs), to characterize their physical properties and to integrate them with superconductor devices and circuits of QC hardware.

The ultimate aim of the project was to investigate genuine single molecules, but since this was not currently feasible, it was decided in the interim to study the quantum state of ensembles of molecules isolated from each other in inert matrices. Two complementary techniques, magnetic susceptibility and magnetic circular dichroism (MCD), were used and it was shown, for the first time, that the magnetic and optical responses are essentially identical. The development of new methods for measurement magnetic and spin properties of isolated molecules is important when the detection limits of traditional SQUID instruments are reached.

The theoretical contribution of the HPLB group (partner 4) was supposed to start on month 6, but due to delays in an appointment, the starting date was shifted to month 12.

The work performed during the second year took into account the modifications to the "Description of work" (Annex 1 to contract) that appeared in the "Revised working plan for years 2 and 3" (June 2003) document.

As far as the preparation of materials is concerned, the work performed was mostly involved with the synthesis and characterization of new  $Mn_{12}$  clusters with interesting optically-active ligands to find synergistic effects. Another important part was the preparation of a series of particles with size and dimensions ranging from a few microns to hundreds of nanometers made from a  $Mn_{12}$  cluster by using new and innovative supercritical fluids techniques. Finally, one of the most important achievements was the successful development of new techniques for the controlled deposition and nanopatterning of  $Mn_{12}$  clusters on a surface over long range distances, by using nanolithographic techniques.

A wide variety of micro-SQUIDs were tested and studied during that year. The NPL group obtained several SQUID from various sources, both from within the consortium and from external contacts which were made during the course of the year and through its co-funding UK DTI activities. A SQUID sensitivity of the order of just 40 spin/Hz<sup>1/2</sup> at 4.2 K was proven by this group. The results were disseminated to other members of the consortium to allow them to improve and enhance their SQUID fabrication. Micro-SQUID fabrication technology was also successfully developed at CSIC-IMM along the second year and operative submicron dimensions devices were processed and distributed for measurements to the partners. A novel approach towards quantum limited spin detection of nanomagnets or magnetic clusters was initiated at CSIC-IMM. Novel nanomechanical devices in GaAs technology promised extraordinary sensitivity and high frequency resonance detection capabilities at low temperatures. First devices were fabricated and tests started.

Most of the efforts concerning microwave measurements focussed on the detection of the coherent radiation appearing after the inversion of population of spin levels in molecular clusters. To carry out this work, InSb bolometers, microwave waveguides, single crystal of molecular clusters, high magnetic fields and low temperatures were used. A cylindrical stainless steel waveguide was first constructed and both the single crystal of molecular clusters with a thermometer and the InSb bolometer were placed inside. The signal of this bolometer was also characterized as a function of temperature, applied magnetic field and current intensity, to allow to estimate the power of detected radiation. The idea of coherent emission (superradiance) was first suggested by R. H. Dicke in 1954, while the idea of the inversion of population in the spin levels of molecular clusters came to us analyzing the data corresponding to the so-called thermal avalanches: at low temperature and high field-sweep rate, sufficiently large single crystals exhibit an abrupt reversal of the magnetic moment, that was attributed to a thermal avalanche in which the initial relaxation of the magnetization towards the direction of the magnetic field results in the release of heat that further accelerates relaxation. In our experiments we detected simultaneously the occurrence of thermal radiation, with the thermometer attached to the single crystal, and the arrival of radiation to the InSb bolometer.

Concerning the qubit measurement, the NPL group successfully imaged  $Mn_{12}$  clusters deposited on various substrate, as prepared by the CSIC group, who visited NPL on three separate occasions. Scanned probe techniques consisting of AFM and MFM as well as room temperature STM and UHV low-temperature STM were employed. Results of AFM and MFM showed clear lines of  $Mn_{12}$  clusters as deposited by the CSIC group. There was strong evidence for a magnetic interaction between the MFM magnetic tip and the individual clusters, suggesting a paramagnetic behaviour at room temperarure. STM investigations were less conclusive and the ability to manipulate  $Mn_{12}$  using STM appeared to be particularly challenging. However, the CSIC group was able to deposit  $Mn_{12}$  clusters within one of the SQUIDs lithographically produced by NPL, and by cooling this device through the  $Mn_{12}$  blocking temperature a change in the SQUID response was seen.

Significant progress towards the theoretical goals laid out in the "Revised working plan for years 2 and 3" document was made by the HPLB group. In particular, the group completed a thorough study of generic qubit plus detector models, using the charge qubit coupled to a point contact detector at arbitrary bias as a canonical example. This work led to a significant discovery: that, contrary to earlier predictions, it is not possible to observe oscillations of an un-driven qubit by continuously monitoring the detector output. In addition, measurement, mixing, relaxation, and dephasing times for this generic model were evaluated for the case when the detector is "on" (large source-drain bias) and "off" (zero source drain bias). The specific case of a current-driven Josephson junction-based measurement device was also studied; this model applies to a DC SQUID being used to measure a nanomagnet or flux qubit, or a DC biased large capacitance junction being used to measure a quantronium qubit. Finally, a preliminary model for a combined nanomagnet-DC SQUID detector was also developed. All this work is extremely relevant for when the experimental work addresses the measurement of a single nanomagnetic qubit using a DC SQUID or related form of detector which couples to the qubit magnetic moment.

#### c. Year 3

The work performed during the third year of this project has been developed as planned in the "Description of work" document (Annex 1 to contract), the "Revised working plan for years 2 and 3" (June 2003) and the appendix to the "Periodic Progress Report No. 2" (January 2004).

With regard to the preparation of molecular materials, the work focused mainly on the improvement of those deposition techniques already established by the CSIC-ICMAB group along the two previous years, combining the use of a polymeric matrix and nanolithographic techniques (WP1, WP3). Another important part was the exploration of new crystallization techniques, all of them based on supercritical fluids, to get a full range of crystal sizes from a few nanometers to microns (WP2). New optically active  $Mn_{12}$  clusters were also synthesised in quest of improved magneto-optical responses (WP2).

Concerning the preparation and investigation of SQUID devices (WP1), CSIC-IMM focused on improving various fabrication steps in order to obtain reliable micro-SQUIDS devices both on niobium (Nb) and aluminium (Al) materials. Introducing a

gold capping and ohmically shunting layer needed special effort due to Al/Au and Nb/Au interdiffusion. NPL tested such devices and also some others provided by CSIRO (Australia) and HYPRES (US). Measurements of the *I-V* characteristics of the devices were made and also for the HYPRES devices small clusters of Mn<sub>12</sub> were deposited within the SQUID loops and the response of the devices measured down to 1 K. Some devices were also imaged using a newly developed technique of SQUID self-portraiture using a magnetically tipped STM system. Devices from CSIC-IMM were tested in both liquid helium cryostats and in a newly commissioned adiabatic demagnetisation refrigerator capable of cooling down to 50 mK. Some of the devices were also sent by NPL to UB for incorporation into a measurement system .

A strong work on the interaction between microwave radiation and molecular magnets was performed by the UB group during this third year. Efforts focused on two main problems: first, the analysis of the electromagnetic radiation accompanying the fast demagnetization process in molecular magnets (WP2, WP3, WP4) and, second, performing new experiments to better understand the spin tunneling transitions in the case of extremely high sweep of the bias magnetic field (WP4, WP6). In fact, these two works connected the behaviour of our magnetic units as qubits and the superradiance emission. Being this phenomenon the most striking effect for decoherence in magnetic qubits, we can assert that the new objectives we were looking for during the last two years turn to be, as a matter of fact, in full relation to those initially proposed.

The experiments to better analyse the electromagnetic radiation coming from molecular magnets were performed using: a) different molecular magnets to correlate the chemical and nuclear properties of the magnetic unit with the probability of emission of radiation (WP2, WP3), b) under different experimental conditions to study the influence of the temperature, magnetic field and number of molecules on the power emission of the electromagnetic radiation (WP4), c) using different waveguides to better correlate the power transmission with the geometry, diameter, length and material, of the waveguides (WP4), and d) using very fast detection methods for the magnetization change and radiation detection (WP4).

The main aim of the experiments performed at ultra high sweep of the bias magnetic field (WP6) was to test the Landau Zener transition probability in the spin tunneling case. It is well establish that single quantum transitions obey the Landau Zener law. In our experiments, however, we have seen that this law is not obeyed anymore and this has been interpreted as due to the simultaneous transition of a huge number of spin levels. This result provides strong evidence for the occurrence of the superradiance emission. These experiments are also crucial to better understand the qubit behaviour of the magnetic clusters when working as logic gates (WP6).

The fundamental physical properties of nanomagnets (WP6) and the development of measurement tools for their investigation (WP5) were further explored by CSIC-IMM and NPL. Patterned arrays of  $Mn_{12}$  clusters prepared by CSIC-IMM were imaged at NPL using a combination of AFM/MFM and ambient STM. Scanned probe methods were also used to try and manipulate small clusters of  $Mn_{12}$  deposited on various substrates as a means of enabling precise  $Mn_{12}$  positioning, as would be the detection of microwave radiation from  $Mn_{12}$  using high- $T_c$  detectors and the performance of magnetisation measurements on  $Mn_{12}$  single crystals. CSIC-IMM contributed with the fabrication of III-V 2D nanostructures for micro-Hall devices and bolometers, and the

fabrication of surface acoustical wave (SAW) devices on LiNbO<sub>2</sub> substrates with modified interdigitated electrodes arrays under demand of UB group. Effort concentrated on optimizing interspace distance and contact geometry for enhanced acoustic wave coupling.

The theory and modelling effort (WP7) delivered basically five pieces of work this year: (i) A study of qubit system driven by external microwaves; (ii) Investigations of various methods of qubit readout (both single shot and weak continuous measurement); (iii) Research on hybrid matter/optical implementations of QIP; (iv) Study of the probing of magnetic qubits with noise; (v) New approaches to detection and gates based on coupling between condensed matter qubits and coherent microwaves. All this work paves the way for the development of experiments on magnetic qubits and the interpretation of the results. (i) and (iv) provide approaches for preliminary investigations of qubits and their environments. (ii) and (v) provide candidate approaches for actual qubit readout for QIP. (v) provides a new approach for quantum gates between qubits, and (iii) provides some new ideas for combined matter/field QIP.

## 2. Project objectives

We have considered appropriate to break down the project objectives in three subsections, one for each year, and a fourth one for the overall management and co-ordination objectives.

#### 2.1. Year 1

The specific objectives for the first year were the following:

- Development of micro-SQUID and Hall effect devices for readout of nanoparticle qubits.
- Preparation and characterisation of magnetic units, both molecular clusters and nanoparticles, to get a full representation of their quantum behaviour, allowing the determination of the experimental conditions to identify qubits.
- Preparation of 2D arrays of single molecule magnets onto a surface by different deposition techniques as well as the manipulation of a single molecule by using magnetic and atomic force microscopy techniques.
- Development of protocols to measure electromagnetic absorption and emission from magnetic samples by using resonant cavities at very low temperature.
- Study of the decoherence processes of the magnetic units, using simulation where necessary.

#### 2.2. Year 2

The specific objectives for the second year were the following:

- Development of the methodology to deposit nanoparticles of Mn<sub>12</sub> molecules, ranging from a micron to a few hundred of nanometers, within a lithographically patterned micro-SQUID.
- $\bullet$  Preparation of new chiral  $Mn_{12}$  magnetic clusters and measurement of their magnetic and optical properties.
- Development of a new methodology to obtain a series of crystals made from  $Mn_{12}$  ranging from a few hundreds of nanometers to a few micra.
- Preparation of very pure materials as well as materials without hydrogen, substituted by deuterium, to modify hyperfine interactions and study decoherence phenomena.
- Improvement of the methodology to address individual single-molecule magnets onto a surface by the use of new lithographic techniques, such as "wetting-dewetting" without a polymeric matrix.
- In-house e-beam lithography and AFM oxidation to produce SQUIDs.
- Development of nano-electro-mechanical devices based on GaAs technology for spin detection.

- Determination of the physical magnitudes involved in the quantum behavior of the magnetic units.
- Simultaneous measurement of the quantum state of the spin system and the emission/absorption of microwaves by the spin levels of molecular clusters and small particles.
- Detection of the emission of coherent radiation by molecular clusters.
- Construction of resonators and use of instrumentation operating until 110 GHz.
- Theoretical analysis and modelling of the measurement process for magnetic qubits, using SQUID magnetometers (or other possible schemes) and taking into account the progress made with the SQUID detectors to date.
- Study of the maximum amount of information about the quantum state (superposition or, for two qubits, entanglement) that can be extracted through weak measurements in a single basis.

#### 2.3. Year 3

The specific objectives for the third year were the following:

- Improvement of those deposition techniques already established by the CSIC-ICMAB group to get better arrangements of Mn<sub>12</sub> clusters on a micro-SQUID environment
- Testing of micro-SQUIDs below 100 mK in order to test the devices in the environment necessary for Mn<sub>12</sub> qubit operation.
- Measurement of SQUID response with Mn<sub>12</sub> cluster during transition through blocking temperature as a means of verifying the device sensitivity and Mn<sub>12</sub> small cluster behaviour.
- Measurement of SQUID spatial sensitivity for nanometre size SQUID to determine the optimum location of any qubit within the SQUID system.
- Synthesis and characterization of new Mn<sub>12</sub> clusters with optically active ligands in quest of improved magneto-optical responses.
- Exploration of new crystallization techniques, all of them based on supercritical fluids, to get a full range of crystal sizes from a few nanometers to microns that should help us to study the influence of factors such as crystal size, polymorphism and crystal defects on the magnetic behavior and relaxation of these clusters.
- Definition and optimization of the measuring set up for low temperature operation of nano-electro-mechanical devices and the corresponding test measurements.
- Full magnetic characterization of the samples prepared.
- Testing of samples with respect to the coupling between the spin system and the different cavity modes.
- Investigation of the correlation between the radiation emission probability with the spin Hamiltonian characterising the magnetic properties of the crystals.

- Investigation of the dependence of superradiance emission on the geometrical arrangements of the quantum dipoles emitting the radiation.
- Distinguishing between thermal black body radiation and the microwave emission of molecular magnets and determining whether this radiation is coherent or not.
- Resolving in time the variation of magnetization and the detection of radiation.
- Spectroscopic studies of superradiance.
- Development of state-of-the-art measurement devices able to interrogate single nano-magnets.
- Coupling of these devices to single nano-magnets to allow the study of the fundamental physical behaviour of the nano-magnets.
- Investigation of the use of noise in a classical resonant (microwave) system as a probe of the quantum state of a nanomagnet qubit or coupled qubits. In particular, investigation of the transport of systems (in a weak measurement scenario) through an avoided crossing of energy levels as signatures of superposition and entanglement.
- Further development of our generic models for continuous quantum measurement, in particular the case of a qubit driven by an external rf-field.
- Development of strategies for quantum measurement in the case where the systemdetector coupling is weak (as expected in the case of nanomagnets coupled to DC SQUIDs).

## 2.4. Management and co-ordination objectives

- Establishment of the necessary flow of information between the partners and coordination of all activities according to the workplan.
- Establishment and maintenance of the contact to the EC.
- Edition of technical progress reports.
- Co-ordination of the activity of each group and integration in the consortium as a whole according to the workplan.
- Administration of the activities of the consortium according to the rules established by the EC.
- Achievement of the best possible dissemination of knowledge.
- Determination of the target groups and the strategic impact of the project.
- Designment of exploitation plans of the results.

## 3. Methodologies

The following methodologies were addressed to fulfill the objectives of the project:

<u>Patterning of  $Mn_{12}$  clusters on a micro-SQUID</u> (WP1). The technique uses a convergent approach based on the stamp-assisted deposition of molecules from a solution together with dewetting phenomena arising from competing interactions between the molecules and the substrate. Measurements to characterize the resulting magnetic particles made from a few hundreds of  $Mn_{12}$  clusters are currently underway. Finally, it has to be emphasized that most of the targets of WP2 were successfully accomplished.

<u>Preparation of magnetic clusters</u> (WP2). The synthetic approach used for the preparation of a variety of derivatives was the ligand substitution reaction, which is driven by the greater acidity of the added carboxylic acids RCO<sub>2</sub>H and/or the removal by distillation of an azeotrope of acetic acid and toluene,

$$[Mn_{12}Ac] + 16 RCO_2H \rightarrow [Mn_{12}O_{12}(O_2CR)_{16}(H_2O)_4] + 16 MeCO_2H$$

where R are different carboxylic groups, like those shown below.

All the complexes obtained following the methodology previously described were chemically characterized by different techniques such as elemental analysis, spectroscopy, MALDI-TOF and X-ray crystallography.

<u>Preparation of nanoparticles</u> (WP2). Nanocrystals of Mn<sub>12</sub> ranging from a few hundred nanometers to one or two micras were obtained using compressed fluids (CF). CF exhibit some very dramatic and attractive solvent properties. Because they are compressible, their density, and subsequently their solvating power, can be controlled with small changes in temperature and pressure. The success or failure of a solution process development (crystallization) is strongly related to the knowledge of the solubility behaviour of the different compounds involved in the process inside the solvent media used. Therefore to be able to design, control and understand processes performed in compressed fluids, it is crucial to know the solubility of the different Mn<sub>12</sub> clusters implicated in the process in the high-pressure solvent mixtures used. In CO<sub>2</sub>-expanded solutions the phenomena of "preferential solvation" or clustering are more enhanced than in solvent mixtures at atmospheric pressure. Therefore small variations in the composition of the CO<sub>2</sub>-expanded solution may cause abrupt changes in the solubility of a given compound, and as a consequence disfavour or enhance a given

process. The phenomena of solvent clustering are highly dependent on molecular solvent-solute and solvent-solvent interactions, and are scarcely studied for CO<sub>2</sub>-expanded solutions.

Magneto-Optic polar Kerr setup (WP2). Samples prepared at CSIC-ICMAB in the form of unsupported polycarbonate films, about 4 μm thick, containing variable concentrations of Mn<sub>12</sub> molecules dissolved in the polymer matrix, were attached on a reflective Si substrate and positioned in the gap of a 1.5 Tesla electromagnet with perforated polar pieces for normal incidence Magneto-Optic polar Kerr effect measurements as a function of wavelength. Such studies were performed by the CSIC-IMM unit. The experimental M-O rotation measuring set-up, for the UV to near IR spectral range (300 to 1000 nm), contains a Xe light source, a grating monochromator, a reflective optics for focusing and guiding the light beam, a polarizing prism, an electro-optic modulator, an analyzer prism and a photomultiplier. Locking detection in phase with the modulator frequency and computer control of the magnetic field sweep and data adquisition completes the set-up which achieves a rotation detection limit of about 10<sup>-3</sup> degrees in the whole spectral range.

<u>Natural circular dichroism</u> (WP2). The chiral nature of the clusters was confirmed by recording their natural circular dichroism (NCD) in dichloromethane at room temperature. All the clusters are optically active, and in contrast to the UV-vis spectra, well defined Cotton effects are observed, but there are important differences in their magnitude and sign. The solid state circular dichroism spectra of ground microcrystals were also recorded in KBr matrix and reveal a pronounced Cotton effect in the visible range at about 550 nm, similar to the ones seen in liquid solution. On the other hand, the Cotton effect at 400 nm is much less intense than that seen in solution.

<u>Preparation of thin-films for controlled deposition of Mn<sub>12</sub> molecules on a surface</u> (WP3). Initially, we developed a new soft, reliable and simple methodology to address individual Mn<sub>12</sub> molecules onto a film surface. Such a methodology is based on the preparation of nanocomposite thin films, made from a polycarbonate polymeric matrix and an Mn<sub>12</sub> complex, and the subsequent treatment with different organic solvent vapours. Solvent treatment originates that a small fraction of the Mn<sub>12</sub> molecules randomly emerge to the surface of the film with an aggregation state that can be controlled at will, depending on the nature of the solvent. This is the first time that small aggregates and individual SMMs are randomly deposited and can be individually screened either by AFM and MFM microscopies. Nevertheless this technique lacks of any control over the orientation and controlled position of the clusters on the surfaces.

For this reason, we improved the methodology by implementing a nanolithographic technique that combines the use of thin-films and stamp-assisted techniques. A solution of polycarbonate (PC) and  $Mn_{12}$  clusters is cast onto a structured master and moulded; upon a solid replica is formed the master is removed. After moulding, the polymer sample is exposed to dichloromethane vapours at room temperature in saturated atmosphere. The solvent vapours partially dissolve the exposed PC surface (the glass-transition temperature is lowered below room temperature) and, due to the requirement of lowering the surface tension, the surface area of the melted polymer is minimised.

The result on topography is the smoothing of the surface features, the surface roughness decreasing linearly with respect to exposure time to the solvent.

Wetting/dewetting for controlled deposition of  $Mn_{12}$  molecules on a surface (WP3). In collaboration with an Italian team, we demonstrated that arrays of nanometer-sized aggregates, each made of a few hundred SMMs derived from  $Mn_{12}$  complexes, can be patterned on large areas by self-organization assisted by stamps on a surface in a dewetting regime. The large length scale is imposed by the motif of the stamp protrusions, and the smaller length scales, viz., the size and distance of the molecular aggregates, are controlled by deposition and growth phenomena occurring in a volume confined beneath the protrusions by capillary forces. The method is general to a variety of molecular materials and substrates because repulsive, as opposed to specific, interactions are required.

<u>Characterisation of SQUID devices</u> (WP1). Standard techniques, encompassing low noise techniques, were used. Some novel plots of the data were devised to aid the analysis of the data. Electromagnetic shielding was employed using standard methodologies for low temperature magnetic measurements. A novel technique of magnetically mapping a SQUID using a magnetically tipped STM was developed giving much greater spatial resolution than more conventional methods such as Hall probe scanning. The methodologies employed are given below:

- (a) SQUIDs were tested in standard 4.3 K non-magnetic helium dewars. The devices were bonded to standard chip holders using a commercial bonder and gold or aluminium wire as needed/appropriate.
- (b) *I-V* curves were recorded as a function of temperature and also of applied magnetic field in some cases. Four wire measurements were always used. Low temperature filter networks and shielding of wires was employed as too was magnetic screening using lead shields.
- (c) Low temperature testing (< 4.2 K) was achieved using an adiabatic demagnetisation refrigerator (ADR). This system is able to cool a mass of 1 kg to 50 mK and is far easier to use than a conventional dilution refrigerator. Special magnetic screening cans were designed and built to house the test devices within the ADR system. Thermometry was achieved using several types of thermometers and several were fixed at key sites within the ADR system.
- (d) The ADR was also fitted with microwave feed throughs so that microwaves can be fed to the test devices in order to show Shapiro steps, for example.
- (e) Field modulation coils were also wound around the ADR sample area so that the magnetic field could be modulated and this effect on the SQUID devices observed.
- (f) The vast majority of data acquisition was achieved through computer control of precision measuring devices. We used the Labview (National Instruments) programming environment for controlling acquisition and display of data.
- (g) All key instruments were calibrated against National Standards where this proved necessary.

*Fabrication of micro-SQUID and micro-Hall probe devices* (WP1, WP5?). Fabrication technology for superconducting micro-SQUID devices and circuits together with technology for micro-Hall probes in III-V heterostructures was set-up and optimised by CSIC-IMM under the feedback of the groups in charge of their characterization. e-beam lithography reliability and speed was substantially improved during the third year by acquiring a new e-beam system (LEO+ Raith) equipped with LaB<sub>6</sub> emitter for much better emission current control and stability. Writing field size was also extended to 500 μm × 500 μm in order to cointegrate and align contact pads with the devices. Positive and negative resist exposure doses and correction software were optimised for the new tool in order to fabricate micro-SQUIDs and micro-Hall probes. All other necessary tools for fabrication as RIE etching, oxide and Si<sub>3</sub>N<sub>4</sub> PECVD deposition, were used throughout the project.

<u>Preparation of surface acoustical wave emitting structures</u> (WP5?). During the last six months of the project a new technology was set under demand of the project coordinator in order to supply surface acoustical wave (SAW) emitting structures on LiNbO<sub>2</sub> substrates for microwave/surface acoustic waves coupling. The CSIC-IMM group implemented design and fabrication technology for SAW devices on LiNbO<sub>2</sub> specially cut piezoelectric sustrates in order to supply specially designed interdigitated arrays to UB for experiments on high frequency microwave excitation of magnetization resonances.

<u>Detection of the emission of electromagnetic radiation by molecular magnets</u> (WP3, WP4, WP6). Two different methodologies were used in the work performed to validate this idea:

#### Methodology 1

- (a) The materials used in our experiments were always a set of single crystals to have in all cases more than 10<sup>18</sup> molecules to cooperate in the coherent decay process. The main idea is the following: the decay process of the spin levels of the molecular magnets is mainly mediated by phonons, as the phonon density is much higher than the photon density. That is, in order to have the emission of radiation, the number of molecules cooperating coherently should be at least as high as the ratio between the transition probability via the emission of photons and phonons.
- (b) The bias magnetic field producing the magnetization/demagnetization process was changed at a speed such to produce the so-called thermal avalanches in the single crystals. The tunneling process at a certain resonant field is thus accompanied by the emission of phonons which excite molecules that tunnel and produce more phonons when decay to the ground state. This very non linear effect produces the population inversion in a time of the order of 1 ms.
- (c) The population inversion is followed by the tunneling of most of the molecules from the same spin level of the metastable well to the same spin level in the stable well.
- (d) This tunneling process is followed by the staircase process to the ground spin state. Experimentally we detect: 1) how much magnetization is tunneling, 2) the time of the duration of the spin tunneling, 3) the temperature change of the sample

- after the thermal avalanche, 4) the time when the emission of radiation occurs, and 5) the power of the emitted radiation.
- (e) The change in the magnetization is measured by pick-up coils surrounding the single crystals. Our coils can detect the magnetization variation with a resolution time better than 1 ms and with a resolution in the magnetic moment better than 10<sup>-4</sup> emu. The temperature of the sample is measured by using two thermomemeters attached to the single crystals with a resolution time of the order of 1 ms at 2 K. The emission of the power of the radiation is measured by using a bolometer placed 13 cm away from the Mn<sub>12</sub> crystals. The set of crystals is electromagnetically coupled to the bolometer by a cylindrical waveguide that may transport electromagnetic radiation above 60 GHz.

#### Methodology 2

In this second methodology we performed experiments in which we studied the dependence of the magnetization reversal of single crystals of Mn<sub>12</sub> and Fe<sub>8</sub> molecular magnets on the sweep rate of the applied magnetic field:

- (a) We used a set of single crystals of  $Mn_{12}$  and  $Fe_8$  molecular magnets having their c-axis aligned with the external magnetic field.
- (b) Our experiments were conducted at temperatures below 1 K and sweep rates of the magnetic field between 10<sup>3</sup> T/s to 10<sup>4</sup> T/s. This very fast field variation was achieved by using pulsed magnetic fields.
- (c) We measured the variation of the magnetization during the fast pulse, so we were capable of detecting simultaneously both the value of the magnetic field at which the magnetic moment reversed and the amount of magnetic moment involved.
- (d) We observed that the systematic shift of the values of the magnetic field at which the magnetization reversal occurred did not obey the law of the Landau-Zener transition model.

<u>Theory and modelling</u> (WP7). This effort delivered basically results in seven general areas. Standard techniques and approaches from general theoretical physics and quantum information were employed:

- (a) Study of the continuous measurement of solid state qubits: Derivation and solution of master equations, decoherence modelled through environment.
- (b) Understanding of the operation of a DC SQUID or large current-biased Josephson junction operating as a qubit measurement system, in particular the knock-on effects to the qubit of the intrinsic irreversibility of a current fed to the measurement apparatus: Lindblad master equation used to model current source. Various mathematical techniques and approximations employed to gain analytic understanding of the system behaviour.
- (c) Understanding of the behaviour of a microwave-driven solid state qubit subject to decoherence the effect of the Rabi oscillation signal on the measurement apparatus: Derivation and solution of master equations, decoherence modelled through environment.

- (d) Study of the readout process (continuous observation and single shot projection) for matter qubits: Derivation and solution of master equations, decoherence modelled through environment.
- (e) Novel proposals for hybrid matter/optical QIP: Application of techniques from quantum optics and quantum information; quantum trajectory approaches used to simulate decoherence effects.
- (f) Study of the probing of magnetic qubits using external thermal noise: Direct adaptation of existing work on quantum effects in SQUID/tank circuit systems.
- (g) New approaches to detection and quantum gates based on the interactions between magnetic qubits and a common microwave bus mode: Application of techniques from quantum optics and quantum information.

## 4. Project results and achievements

#### WP1. Micro-SQUID and Hall probe development

Molecular cluster thin-films deposited on SQUID and patterned by lithography

The CSIC-ICMAB unit achieved the patterning of aggregates of  $Mn_{12}$  SMMs on a lithographically patterned micro-SQUID provided by the NPL group. The technique uses a convergent approach based on the stamp-assisted deposition of molecules from a solution together with dewetting phenomena arising from competing interactions between the molecules and the substrate (vide infra). Measurements to characterize the resulting magnetic particles made from a few hundreds of  $Mn_{12}$  clusters were performed by NPL.

#### Micro-SQUID development

Objectives of the CSIC-IMM unit within the project were initially limited to supply fabrication technology for superconducting devices, such as state-of-the-art micro-SQUIDs and superconducting circuits and switches for magnetic qbit preparation and characterization. Evolution of the project forced the activity of this unit to focus on extremely small micro-SQUID development, trying to extend the device sensitivity beyond state-of-the-art and to be able to measure single spin clusters with S=10. Interest on this extremely difficult to achieve objective with CSIC-IMM experience and technology faded out as measurement attempts at NPL and contacting difficulties resulted in blown-up or short-circuit devices at low temperatures.

The experience gained through these developments is of great value for our scientific community and society. First, because this project has directly promoted the establishment of the first operative e-beam nanofabrication laboratory in our country and helped training technical personnel in a field previously absent. Second, because this know-how has effectively spread to other two labs in Spain (UAM-CSIC and U Zaragoza) using e-beam lithography at present. Third, because the competitiveness of the fabrication technology at IMM has been enhanced to the point in which, for the first time, it is possible to start competing in high tech projects for the ESA (Superconducting Bolometers for X-Ray Detection and Spectrometry). Synergies with other relevant projects within the group such as IST-NANOMAT and recently SANDIE NoE have been also very positive, as extending the range of applications to the III-V compound nanostructures.

NPL tested many and developed several micro-SQUID devices. Both aluminium and niobium based devices from CSIC-IMM, and devices obtained from commercial fabrication facilities (HYPRES in US) and from world leading research groups (Wernsdorfer in Greoble, Lam, CSIRO in Australia) were tested. NPL also sent some devices to UB for inclusion in their experiments on microwave emission from Mn<sub>12</sub> single crystals. The sensitivity of the devices was measured and a full characterisation of their *I-V* curves as functions of temperature and their response to magnetic fields was performed. Using a newly aquired apparatus for this project, an adiabatic demagnetisation refrigerator, NPL were able to test devices down to 50 mK simulating the operating conditions of the devices if they are to be used as qubit detectors. Using

our measurements we were able to give feedback to the fabrication process so that better devices could be made. An example was the recommendation that a gold overlayer of specific thichkness be applied to the niobium devices from CSIRO and that the geometry of the junction design was changed. This work enabled us to form relations and synergies with these important groups and so to raise our profile and capability within Europe. These devices have potentially a wide range of applications that can impinge on various aspects of the quality of life, for example as THz detectors for security screening.

#### WP2. Qubit preparation, identification and characterisation

Preparation of new magnetic clusters and nanoparticles and measurement of their magnetic properties.

Molecular clusters. Complex  $[Mn_{12}O_{12}(O_2CC_6H_4N(O)^tBu)_{16}(H_2O)_4]$ , bearing openshell radical units, were prepared by CSIC-ICMAB. The expectation was that the radical ligands may couple ferromagnetically with the  $Mn_{12}$  core. Unfortunately, magnetic measurements performed by the UB group revealed that the sixteen radical carboxylate ligands interact antiferromagnetically with the  $Mn_{12}$  core to yield a S=2 magnetic ground state. Moreover, three chiral dodecamanganese clusters that behave as SMMs and reveal natural optical activity, were prepared. These new chiral SMMs, the first of their kind, also reveal optical bistability when monitored using MCD. The natural and magnetic circular dichroic properties of the isolated molecules in dichloromethane—toluene glass bode well for the observation of magneto-chiral dichroism in these SMMs under more sensitive experimental conditions.

<u>Nanoparticles</u>. Nanocrystals of  $Mn_{12}$  ranging from a few hundred nanometers to one or two microns were obtained by using supercritical fluids. Different crystallization techniques, such as GAS and ASES, were used. Among them, GAS proved to be the most effective for the preparation of nanocrystals with quite a monodispersize size, and with different size and shape, both of them controlled at will.

Determination of the physical magnitudes involved in the quantum behavior of the magnetic units.

Kerr Effect. Initial set of measurements in collaboration with CSIC-IMM were intended to determine the sensitivity of the method and the adequacy of the sample preparation method. Signal amplitude is quite small in the measured range, due to the limited magnetic field intensity achievable and the super-paramagnetic nature of the samples but, in principle, useful to determine optical transitions and their magnetic dependence. Systematic measurements as a function of temperature in a low temperature cryostat and sample preparation conditions, are envisaged in the next future. As far as we know, this is the first effort devoted to the observation of Kerr Effect on SMMs and the technical implications that can be derived from there.

<u>Magneto-optical spectroscopy (Magnetic Circular Dichroism).</u> Magnetic and magneto-optical measurements of Mn<sub>12</sub> SMMs with no-chiral ligands in organic solvent glasses were carried out by CSIC-ICMAB in close collaboration with UB. Magnetic

characterization of these samples by SQUID magnetometry contrasts with that of microcrystalline samples in that the relative proportions of the two relaxation processes active in these compounds are altered. Field-dependent magnetic circular dichroism (MCD) intensity curves (hysteresis cycles) are found to be essentially identical to the SQUID results and provide experimental evidence for the potential of the optical technique for magnetic characterization. Indeed, since the MCD signal is directly proportional to magnetization measurements performed on  $Mn_{12}$  SMMs at visible wavelengths, this permitted the determination of properties from blocking temperatures to the degrees of quantum coherence and superposition of states. The technique was also used in preliminary investigations of the influence of the environment on these SMMs, finding that the  $Mn_{12}$  systems isolated in glassy media show a greater preponderance of fast-relaxation species, especially in the case of  $CH_3CN$ :dmf.

Finally, optical observation of magnetic tunneling was achieved for the first time by studying the decay of the MCD signal at weak applied magnetic fields. In addition, three chiral dodecamanganese clusters (bearing chiral ligands) that behave as SMMs and reveal natural optical activity, were prepared. For the cluster with 2-chloropropionate moieties at its periphery, the magnetic circular dichroism was investigated and found to display large optical hysteresis, which depends on the direction in which the magnetic field direction is swept. The use of enantiopure 2-chloropropionate groups as the peripheral ligands endow appreciable natural optical activity upon the dodecamanganese core that they surround. This feature is very dependent on the particular chiral ligand, since neither of the chiral (S)-6-methoxy-amethyl-2-naphthaleneacetate nor the (S)-2-phenylbutyric acetate compounds reveal such significant Cotton effects in their CD spectra.

#### WP3. Qubit deposition and purification

Controlled deposition of magnetic qubits in pure quantum spin states

Approach 1. The CSIC-ICMAB unit developed a new soft, reliable and simple methodology to address individual  $Mn_{12}$  molecules onto a film surface. Such a methodology is based on the preparation of nanocomposite thin films, made from a polycarbonate polymeric matrix and a  $Mn_{12}$  complex, and the subsequent treatment with different organic solvent vapours. Solvent treatment originates that a small fraction of the  $Mn_{12}$  molecules emerge to the surface of the film with an aggregation state that can be controlled at will, depending on the nature of the solvent. Moreover, we shown that the location of the molecules on the surface can be directed by the use of lithographic techniques and the use of a stamp. Indeed, such a methodology is based on the preparation of a polymeric thin-film made from a polycarbonate matrix and  $Mn_{12}$  molecules, which is moulded with a master and exposed to vapours of an organic solvent. Such a solvent treatment smooths out the surfaces relieves of the polymeric replica and the  $Mn_{12}$  molecules that were dispersed in the protruding regions of the replica concentrate in the original protruding features but do not diffuse laterally, thus originating a compositional contrast of  $Mn_{12}$  on the polymeric surface.

Magnetic response of the patterned aggregates of  $Mn_{12}$  molecules, using Magnetic Force Microscopy (MFM), follows accurately the compositional spatial modulation of the pattern, so it is possible to encode and read out information with such molecular patterns working at the paramagnetic regime from mesoscopic down to nanometer

length scales. This is the first time that small aggregates and individual SMMs are deposited and can be individually screened either by AFM and MFM microscopies. The advantages of this approach are considerable. First, the polymeric matrix plays a critical role in stabilizing the SMM as well as enhancing the overall mechanical strength of the film. Second, polycarbonates are a commercially important and technologically interesting polymers that have a combination of properties not found in any other plastic, including very high impact strength, creep resistance, optical clarity, and a low moisture absorption. And finally, the resulting nanocomposite thin films are fully compatible with nowadays magnetic and magneto-optical storage technologies, where polycarbonate resins are used as the basic support for the fabrication of disks.

Approach 2. Along the second year, in collaboration with an Italian team, the CSIC-ICMAB unit demonstrated that arrays of nanometer-sized aggregates, each made of a few hundred single-molecule magnets derived from Mn<sub>12</sub> complexes, can be patterned on large areas by self-organization assisted by stamps on a surface in a dewetting regime. The large length scale is imposed by the motif of the stamp protrusions, and the smaller length scales, viz., the size and distance of the molecular aggregates, are controlled by deposition and growth phenomena occurring in a volume confined beneath the protrusions by capillary forces. The method is general to a variety of molecular materials and substrates because repulsive, as opposed to specific, interactions are required. Our result hints at the possibility of sustainable patterning of single-molecule magnets for ultra-high-density magnetic storage and quantum computing.

Purification and isotope enrichment of magnetic qubits

Different deuterated samples of the well-known  $Mn_{12}Ac$  ([ $Mn_{12}O_{12}(CD_3COO)_{16}$  ( $D_2O)_4$ ].2 $CD_3COOD.4H_2O$ ) were obtained following the methodology reported in the literature by Lis for the synthesis of  $Mn_{12}Ac$  by using deuterated acetic acid and  $D_2O$ . Characterization of their magnetic properties and resonant experiments under microwave radiation was done by the UB group.

#### WP4. Microwave and RF measurements

Concerning the results obtained in the field of microwave radiation, we should begin mentioning that the search of microwave superradiance was first ignited inside the current project. Due to the work on the search of the magnetic qubits we came to the idea that superradiance emission could interfere with the operation of these qubits as logic quantum gates, as it would determine entirely the time of their operation. Our findings of the fast reversal of the magnetic moment when looking for the fast magnetic response of the magnetic qubits was the signal that superradiance could be present in our experiments. As the project developed, the research in this field spread to cover the other workpackages the UB group was involved in (WP2, WP3, WP6). We summarise here the evolution of the ideas and the main results achieved in

Our first experiments to capture the effect of the spin quantum decoherence were performed by observing the <u>electromagnetic absorption of a very pure and well characterised single crystal of  $Mn_{12}$  introduced in a resonant cavity. Two main results came out from these experiments:</u>

- 1. The quantum relaxation due to the quantum tunneling spin transitions are modified when the molecules are inside the cavity. That is, the electromagnetic modes of the cavity affect the quantum spin transitions, and we suggested that this effect could be related to the fact that the staircase transitions to the ground state occurring after the spin tunneling transitions may generate electromagnetic radiation.
- 2. The spin state of the molecules inside the cavity depends strongly on the resonant frequencies of both the cavity and the waveguides used in the experiments. Our main idea was the following: Let us call  $|1\rangle$  and  $|0\rangle$  the two states between which the spin oscillates and consider the system is prepared in the arbitrary superposition  $|\Psi\rangle = C_1 |0\rangle + C_2 |1\rangle$ , in which the spin oscillates coherently between its two orientations at the frequency  $\omega = \Delta/\hbar$ , where  $\Delta$  is the energy difference between the two initial states. Interaction of the above quantum states with the dissipative environment results in the decoherence damping of the oscillations, due to, *e.g.*, the decay of  $|1\rangle$  into  $|0\rangle$ , which limits the quality factor of the qubit. Our main conclusion from the above mentioned work is that for an ensemble of closely-packed identical qubits, the Dicke superradiance must be the dominant mechanism of decoherence.

The next step of our work was to <u>detect the radiation emitted from molecular magnets</u>. We tried this by performing two completely different experimental procedures which can be distinguished by the bias field sweep rate: in the first one it was small, of the order of 10 Oe/s, while in the second one it was enormous, of the order of  $10^3$  Oe/s to  $10^4$  Oe/s. In the first case, the main idea was to produce the following chain of events:

- 1. Population inversion in the spin states in the metastable well.
- 2. Simultaneous spin tunneling transitions to the corresponding spin level at the stable well.
- 3. Coherent spin transitions form this level to the immediate lower spin level.
- 4. Detection, in case of emission, of the electromagnetic radiation coming out from these transitions involving a huge number of spins (of the order of 10<sup>18</sup> molecules).

We strongly believe that our work in this field is pioneering this new research area and we have accumulated enough expertise in Europe to lead the field in the next years. It is also important to remark that all the knowledge generated can be, in principle, easily projected onto the search of new industrial applications. In this scope it is worth to mention that in case of generating a mW power of microwave radiation from our molecular magnets, we will have in hand the most powerful emitters of coherent radiation. In relation to the fields in which coherent microwave radiation may be used, it is worth mentioning medicine and biology as new tools for identifying molecules and agglomerates of molecules and security as detector of objects and materials. In all these three fields, the EC is very much interested and has asked for standards.

It is also worth mentioning the relations and synergies of this project with a similar project the UB group is developing in collaboration with the Spanish Administration and several Spanish industries in the field of the possible uses of microwave radiation in a broad spectrum of industrial applications, such as the use of molecular magnets as very sensitive and very frequency dependent bolometers, or the use of molecular magnets as a new source for microwave radiation. The work of the UB group within the current project was very much helped by these collaborations, particularly because it

was possible to buy very new microwave network analyzers and spectrometers as well as several bolometers and radiation detectors.

#### WP5. Development of measurement tools for quantum nano-magnets

Mn<sub>12</sub> patterned arrays supplied by CSIC were imaged by NPL using scanned probes ranging from AFM, MFM STM to UHV-Low temperature STM. The imaging of magnetic nanoparticles by scanned probes as we demonstrated is also a vital aspect for many of the areas of emerging nano-technology; for example, this work impacts on data storage devices and new magnetic storage medium based on magnetic nano particles. The manipulation of nano-particles needed for qubit preparation is also applicable to a wide variety of applications where a bottom-up device fabrication route is needed. The work of NPL in this area also lead to collaborations and interactions with the University of Posrtsmouth where we used the techniques learnt in this project to image and manipulate magnetic beads attached to DNA. An aspect that arose is that the measurement of magnetism on a nanoscale is both qualitative and non-traceable at the present. If future claims on magnetic properties of nanoparticles is to be verified, then work is needed to develop methods and standards that can address this problem. NPL and its European partners are well placed to take up this challenge.

On the other hand, as the project developed, interest shifted into microwave superradiance emission and detection in magnetic assemblies along the third year, and CSIC-IMM was asked to stop in-house development of more promising small magnetic moment detection by nano-electro-mechanical devices fabricated on GaAs and to supply tools for microwave detection and surface acoustical wave (SAW) devices.

#### WP6. Exploration of the fundamentals of quantum nanomagnets

NPL masked magnetisation curves on several samples of  $Mn_{12}$  single crystals supplied by CSIC. These measurements were performed using a SQUID magnetometer/susceptometer system at Oxford University allowing to form a close working relationship with their group. In collaboration with Chalmers University (Sweden), NPL also incorporated novel high temperature superconductor Josephson junction detectors. This link to a foremost European institute is proving very beneficial to both parties. Combining the measurement expertise at NPL with the fabrication facilities at Chalmers puts Europe in a very strong position to lead future work in the THz detection and source areas which may well have a very important impact on future quality of life.

#### WP7. Theory and simulations

As regards the theory and modelling contribution to the project, in comparison with the original objectives, we have addressed the problems we set out to consider, but have also widened our horizons to consider some other important aspects. A deliberate feature of the approaches we have used is to make our theory and modelling as generic as possible. This means, for example, that our detector-environment-measurement can be adapted and applied to a number of solid state scenarios (magnetic qubit coupled to SQUID, electron qubit coupled to QPC, charge qubit coupled to SET, etc.). As a

consequence, much of the work we have done, in addition to being highly relevant for future experiments on magnetic qubits probed by SQUID systems (the original Nanomagiqc objective), is also relevant to numerous other investigations being made worldwide on various forms of solid state qubits and their measurement. This is reflected in the various references cited below in the descriptions of the work, the coauthors on some of our papers being outside Nanomagiqc, and the fact that a number of our papers are in general-interest, widely-read physics journals.

## 5. Deliverables and references

#### 5.1. Deliverables

## a. Description of deliverables

D1.1 (Month 24): Report on the applicability of micro-SQUID and micro-Hall probes to the measurement of the properties of nanomagnets.

A comparison of the characteristics of the range of state-of-the-art probes that can be used to measure magnetic moments and their dynamic properties of isolated nanomagnets at low temperatures.

D1.2 (Months 24, 36): Report on measurement devices and methods to measure the properties of small clusters of nanomagnets having total spin of 1000 or less and the suitability of such methods to the measurement of single magnetic qubits.

The range of devices considered within the project was increased by the addition of state-of-the-art nano-electro-mechanical devices being developed by CSIC-IMM. State-of-the-art nanoscale SQUIDs are being tested at NPL. Initial coupling of nano-magnets to nano-SQUIDs was achieved.

In the third year, measurements were made on small clusters of nanomagnets that contained approximately 100 molecules, thus the total spin was of the order of 100 or less. The magnets were flip-chipped to the micro-SQUIDs and then cooled to approx 1 K and the SQUID behaviour recorded.

D2.1 (Month 24): Report on qubit preparation, identification and characterisation.

New chiral nanomagnets with interesting magneto-optical properties were synthesized and a new technique to obtain crystals with variable sizes, ranging from a few hundreds of nanometers to a few micra, was developed. In addition, a new experimental protocol consisting of the combination of both magnetic methods and cavity experiments to fully characterize the spin state of molecular magnets was followed.

D3.1 (Month 24): Report on the findings of the deposition of the magnetic qubits in pure quantum states on different matrices and substrates.

We have developed a new technique to obtain arrays of nanometer-sized aggregates, each made of a few hundred single-molecule magnets, patterned on large areas on a thin-film or different substrates (HOPG, SiO<sub>2</sub>, etc.) by self-organization assisted by stamps.

D3.2 (Months 24, 36): Report on the purification of magnetic units as qubits.

Different deuterated samples of the well-known  $Mn_{12}Ac$  ([ $Mn_{12}O_{12}(CD_3COO)_{16}$  ( $D_2O)_4$ ].2 $CD_3COOD.4H_2O$ ) were obtained following the methodology reported in the literature by Lis for the synthesis of  $Mn_{12}Ac$  by using deuterated acetic acid and  $D_2O$ . With this isotopic enriched pure complex, that could be used as qubits, single crystals of different sizes for future physical measurements were also obtained.

The purification of the samples used for the superradiance experiments was an essential process. The apperance of superradiance emission as a huge collective manifestation of

the simultaneous decay of an ensemble of molecular magnets between spin states require mostly two ingredients: (1) the existence of an interaction between all the quantum dipoles decaying, and (2) a well determined geometry of the molecules inside the sample of a millimetric size. Our observation of the dependence of the power of the radiation detected on the purity and single crystals arrangements may well be the evidence of this cooperative effect.

D4.1 (Month 6): Report on the findings of the study of the resonant system and prototypes of working cavities, fixtures and sample holders.

The report on the study of the equipments needed for the observables to be measured (power absorption, power emission and resonant frequencies) at frequencies up to hundred GHz was included in the first yearly periodic progress report.

Cavities, fixtures and sample holders for resonant experiments were developed during the first year:

- Cylindrical cavities of different diameter and adjustable length and a coaxial cavity of fixed length were constructed using 99.99-%-pure copper.
- Fixture and sample holders were also made using 99.99-%-pure copper.
- Resonant loops of different diameter were constructed using either 0.1-mm-thick 99.99-%-pure copper or 0.015-mm-thick superconducting niobium-titanium alloy.

D4.2 (Month 12): Report on Microwave and RF measurements.

Brief report on microwave and RF measurements describing the work performed in workpackage WP4 during the first year.

D4.3-D4.4 (Months 24, 36): Report on the correlation between the quality factor and the isotope enrichment and interaction between qubits and environment.

A new method was developed to determine the quality factor of microwave resonators containing magnetic materials based upon the modifications of the magnetization of the material at the resonant frequencies. This work was done using different resonators, like coils and cavities of different shape, and different magnetic materials like molecular clusters, high temperature superconductors, ferromagnetic materials and paramagnets.

Measurements were made on deuterated single crystals of  $Mn_{12}$  acetate to study the influence of the absence of nuclear spin in both the emission of electromagnetic radiation and the sharpness of the spin tunneling transitions. The idea is simple: by decreasing the number of nuclear spins surrounding the electronic spin, less number of hyperfine states will be formed and one may consequently expect to reduce the decoherence phenomena. Although we have not be able to quantify the cause–effect relation between radiation power and purity of the sample, we may say that our results suggest the existence of a strong correlation between them.

D5.1 (Month 30): Report on the applicability of nano-SQUIDs and other devices to measure the properties of nano-magnets having spins of 1000 or less.

Sensitivity measurements on micro-SQUIDs show that their intrinsic sensitivity is sufficient for them to be a realistic detection candidates for Mn12 based qubits. Scaling down of SQUID loop size to sub-micron is also possible using e-beam lithography and the technique should allow for easy scaling to large arrays of devices.

D5.2 (Month 30): Update report on nano-magnet measurement devices.

NPL measured and investigated SQUID based devices. These ranged from Nb devices to Al devices and in size from a few microns down to 200 nm loop dimensions. Sensitivity and applicability to  $Mn_{12}$  spin detection were addressed.

D5.3 (Month 36): Report on using devices as qubit readout devices.

NPL addressed the applicability of nano-SQUIDs as qubit readout devices in terms of their operating conditions, scalability of the fabrication technology and practical sensitivity.

D6.1 (Month 24): Report on the study of measurement system.

The fundamentals and chances to fabricate nano-electro-mechanical devices with sufficient sensitivity to measure the spin state of nanomagnets were explored. The necessary know-how to allow us to establish a robust coupling between the spin system and the electromagnetic modes of microwave resonators, allowing to better determine the magnetic state of the magnetic material, was developed. A review of the scarce and novel publications and the state-of-the-art of the devices was included.

D6.2 (Month 36): Report on measurements of fundamental properties of nano-magnets and their future applicability to QIP.

Measurements of the magnetisation of single crystals of  $Mn_{12}$  as a function of temperature and magnetic field were made by NPL using a SQUID susceptometer at Oxford University, UK. Also NPL set up a measurement method to detect the microwave radiation emanating from single crystals using novel high temperature superconductor Josephson junctions.

In addition, two different types of experiments looking for radiation emission were performed by UB. In the first type of experiments, the radiation was directly detected by using bolometers and transporting the radiation using waveguides. From these experiments the power emission was correlated to the different parameters governing the emission of superradiance in general. In the second type of experiments, the tunneling transition for fast sweep rate was found to be in agreement with the emission of superradiance and not longer governed by the Landau Zener law.

D7.1 (Month 36, originally intended for Month 30): Reports/papers on the investigations of the objectives.

A number of theoretical papers on the work performed for this project were produced, and others are in preparation.

D7.2 (Throughout project): Constructive feedback and appropriate theoretical support to the experimental investigations, to enable the thorough investigation of magnetic systems against the required criteria for QC hardware.

The attention was focused on the interaction between qubits and measurement apparatus, with particular emphasis on the decohering effects of the measurement system on the qubit. These results are important for assessing the likely outcomes of experimental measurements on nanomagnet qubits. In year 3, the quantum theory of SQUID measurement, a theoretical description of continuous measurement of a qubit undergoing Rabi oscillations and an approach to interacting and measuring qubits

through a common microwave "bus" mode were further developed. All this work, combined with that from year 2 paves the way for the future understanding of experimental results on magnetic qubits coupled continuously to measurement apparatus such as SQUIDs and driven with microwave fields. It also provides theoretical support for the design and subsequent refinement of such experiments.

D8.1 (Month 6): Web page set.

Public access to a project website (http://www.nanomagiqc.com) is available since the beginning of July 2002.

D8.2 (Month 6): Project presentation.

This presentation appeared in the website on month 12. The presentation helps the non-specialist to get an insight into the project at a glance by listing the participants, the total cost and the commission funding, and briefly describing the main goals, the key issues, the technical approach and the expected impact.

D8.3-D8.4 (Month 12): First year periodic progress report.

Supplied to the Commission on month 12.

D8.5-D8.6 (Month 24): Second year periodic progress report.

Supplied to the Commission on month 24.

D8.7 (Month 36): Third year periodic progress report.

Supplied to the Commission on month 36.

D8.8 (Month 36): Final report.

Supplied to the Commission after month 36, at the final review meeting.

D9.1 (Month 6): Dissemination and Use Plan.

Supplied to the Commission on month 24. Describes plans for the dissemination of knowledge gained during the project and tentative plans for the exploitation of the results achieved.

D9.2 (Month 12): List of Publications.

Supplied to the Commission after month 12, at the review meeting.

D9.3 (Month 24): List of Publications.

Supplied to the Commission after month 24, at the review meeting.

D9.4 (Month 36): List of Publications.

Supplied to the Commission after month 36, at the final review meeting.

D9.5 (Month 36): Technological Implementation Plan.

Supplied to the Commission after month 36, at the final review meeting.

# b. Deliverables table

Del. No.	Title	Type <sup>1</sup>	Classification <sup>2</sup>	Due Date	Issue Date
D1.1	Report on the applicability of micro-SQUID and micro-Hall probes to the measurement of the properties of nanomagnets	R	Pub	24	24
D1.2	Report on measurement devices and methods to measure properties of small clusters of nanomagnets	R	Pub	24, 36	24, 36
D2.1	Report on qubit preparation, identification and characterisation	R	Pub	24	24
D3.1	Report on the findings of the deposition of the magnetic qubits in pure quantum states on different matrices and substrates	R	Pub	24	24
D3.2	Report on the purification of magnetic units as qubits	R	Pub	24, 36	24, 36
D4.1	Cavities and fixtures	O*	Pub	6	12
D4.2	Report on microwave and RF measurements	R	Pub	12	12
D4.3-D4.4	Report on the correlation between the quality factor and the isotope enrichment and the interaction between qubits and the environment	R	Pub	24, 36	24, 36
D5.1	Report on the applicability of nano-SQUIDs and other devices to measure the properties of nano-magnets having spins of 1000 or less	R	Pub	30	36
D5.2	Update report on nano-magnet measurement devices	R	Pub	30	36
D5.3	Report on using devices as qubit readout devices	R	Pub	36	36
D6.1	Report on the study of the measurement system	R	Pub	24	24
D6.2	Report on measurements of fundamental properties of nano-magnets and their future applicability to QIP	R	Pub	36	36
D7.1	Reports/papers on the investigations of the objectives of WP7	R	Pub	36	36
D7.2	Constructive feedback and appropriate theoretical support to the experimental investigations	O**	Int	Throughout	Throughout

D8.1	Website	O***	Pub	6	6
D8.2	Project presentation	O***	Pub	6	12
D8.3-D8-4	First year periodic progress report	R	FP5	12	12
D8.5-D8.6	Second year periodic progress report	R	FP5	24	24
D8.7	Third year periodic progress report	R	FP5	36	36
D8.8	Final report	R	FP5	36	36
D9.1	Dissemination and Use Plan	R	Pub	6	24
D9.2	List of publications	R	Pub	12	12
D9.3	List of publications	R	Pub	24	24
D9.4	List of publications	R	Pub	36	36
D9.5	Technological Implementation Plan	R	Pub	36	36

<sup>\*</sup>Prototype; \*\*Communication within the Consortium, \*\*\*http://www.nanomagiqc.com

IST: Circulation within IST Programme participants
FP5: Circulation within Framework Programme participants

Pub.: Public documen

<sup>&</sup>lt;sup>1</sup> R: Report; D: Demonstrator; S: Software; W: Workshop; O: Other – Specify in footnote

<sup>&</sup>lt;sup>2</sup> Int.: Internal circulation within project (and Commission Project Officer + reviewers if requested)
Rest.: Restricted circulation list (specify in footnote) and Commission SO + reviewers only

## 5.2. References

## a. Articles

- R. Amigó, E. del Barco, Ll. Casas, E. Molins, J. Tejada, I. B. Rutel, B. Mommouton, N. Dalal, J. Brooks, "Quadratic Transverse Anisotropy Term due to Dislocations in Mn<sub>12</sub> Acetate Crystals Directly Observed by EPR Spectroscopy", *Physical Review B* 65 (2002) 172403.
- J. M. Hernandez, F. Torres, J. Tejada, E. Molins, "Crystal Defects and Spin Tunneling in Single Crystals of Mn<sub>12</sub> Clusters", *Physical Review B* **66** (2002) 161407(R).
- J. Gallop, P. W. Josephs-Franks, J. Davies, L. Hao, J. Macfarlane, "Miniature dc SQUID devices for the detection of single atomic spin-flips", *Physica C* **368** (2002) 109-113.
- R. Amigó, J. M. Hernandez, A. García-Santiago, J. Tejada, "High-resolution detection of resonant frequencies of microwave resonators via magnetic measurements", *Applied Physics Letters* **82** (2003) 4528-4530.
- R. Amigó, J. M. Hernandez, A. García-Santiago, J. Tejada, "Microwave absorption and magnetization tunnelling in Mn<sub>12</sub>-acetate molecular clusters", *Physical Review B* **67** (2003) 220402(R).
- R. Amigó, J. M. Hernandez, A. García-Santiago, J. Tejada, "Magnetic detection of millimeter waves", *Europhysics Letters* **64** (2003) 158-163.
- J. Tejada, R. Amigó, J. M. Hernandez, E. M. Chudnovsky, "Quantum dynamics of crystals of molecular nanomagnets inside a resonant cavity", *Physical Review B* 68 (2003) 014431.
- D. Ruiz-Molina, M. Mas-Torrent, A. I. Balana, N. Domingo, J. Tejada, M. T. Martínez, C. Rovira, J. Veciana, "Isolated Single-Molecule Magnets on the Surface of a Polymeric Thin Film", *Adv. Mater.* **15** (2003) 42.
- P. Gerbier, D. Ruiz-Molina, N. Domingo, D. B. Amabilino, J. Vidal-Gancedo, J. Tejada, J. Veciana, "Synthesis and Characterization of a [Mn<sub>12</sub>O<sub>12</sub>(O<sub>2</sub>CR)<sub>16</sub>(H<sub>2</sub>O)<sub>4</sub>] Complex Bearing Paramagnetic Carboxylate Ligands. Use of a Modified Acid Replacement Synthetic Approach", *Monatshefte für Chemie-Chemical Monthly* **134** (2003) 265.
- N. Domingo, P. Gerbier, J. Gómez, D. Ruiz-Molina, D. B. Amabilino, J. Tejada, J. Veciana, "Synthesis and characterization of a new chiral nanomagnet", *Polyhedron* 22 (2003) 2355.
- D. Ruiz-Molina, M. Mas-Torrent, A. I. Balana, N. Domingo, J. Tejada, M. T. Martínez, C. Rovira, J. Veciana, "Single-molecule magnets on a polymeric thin film as magnetic quantum bits", *SPIE Proceedings* **5118** (2003) 594.
- N. Domingo, B. E. Williamson, J. Gómez-Segura, Ph. Gerbier, D. Ruiz-Molina,
   D.B. Amabilino, J. Veciana, J. Tejada, "Magnetism of Isolated Mn<sub>12</sub> Single-molecule Magnets Detected by Magnetic Circular Dichroism: Observation of Spin

- Tunneling with a Magneto-optical Technique", *Physical Review B* **69** (2004) 052405.
- L. Hao, J. C. Gallop, C. Gardiner, P. Josephs-Franks, J. C. Macfarlane, S. K. H. Lam, C. Foley, "Inductive superconducting transition-edge detector for single-photon and macro-molecule detection", *Superconductor Science and Technology* **16** (2003) 1479-1482.
- P. W. Josephs-Franks, L. Hao, A. Tzalenchuk, J. Davies, O. Kazakova, J. C. Gallop, L. Brown, J. C. Macfarlane, "Measurement of the spatial sensitivity of miniature SQUIDs using magnetic-tipped STM", Superconductor Science and Technology 16 (2003) 1570-1574.
- J. C. Gallop, "SQUIDs: some limits to measurement", *Superconductor Science and Technology* **16** (2003) 1575-1582.
- L. Hao, J. C. Macfarlane, P. W. Josephs-Franks, J. C. Gallop, "Inductive superconducting transition edge photon and particle detector", *IEEE Transactions on Applied superconductivity* **13** (2003) 622-625.
- P. Gerbier, D. Ruiz-Molina, J. Gómez, K. Wurst, J. Veciana, "Examining Thermolysis Reactions of Nanoscopic Mn<sub>12</sub> Single Molecule Magnets", *Polyhedron* 22 (2003) 1951.
- M. Cavallini, F. Biscarini, J. Gómez, D. Ruiz-Molina, J. Veciana, "Multiple length scale patterning of single-molecule magnets", *Nano Letters* **3** (2003) 1527.
- B. Schelpe, A. Kent, W. Munro, T. Spiller, "Inferring superposition and entanglement from measurements in a single basis", *Phys. Rev. A* **67** (2003) 052316.
- M. Jordi, A. Hernandez-Mínguez, J. M. Hernandez, J. Tejada, S. Stroobants, J. Vanacken and V. V. Moshchalkov, "Scaling of the susceptibility vs. magnetic-field sweep rate in molecular magnet", *Europhys. Lett.* **68** (2004) 888.
- J. Vanacken, S. Stroobants, M. Malfait, V. V. Moshchalkov, M. Jordi, J. Tejada, R. Amigó, E. M. Chudnovsky, D. Garanin, "Pulsed field studies of the magnetization reversal in molecular nanomagnets", *Phys. Rev B* **70** (2004) 220401.
- Ph. Gerbier, N. Domingo, J. Gómez, D. Ruiz-Molina, D. B. Amabilino, J. Tejada, B. E. Williamson, J. Veciana, "Chiral, Single-Molecule Nanomagnets: Synthesis, Magnetic Characterization and Natural and Magnetic Circular Dichroism", *J. Mater. Chem.* 14, 2455 (2004).
- T. M. Stace and S. D. Barrett, "Continuous quantum measurement: inelastic tunnelling suppresses current oscillations", *Phys. Rev. Lett.* 92, 136802 (2004).
- A. Nazir, B. W. Lovett, S. D. Barrett, T. P. Spiller, and G. A. D. Briggs, "Selective spin coupling through a single exciton", *Phys. Rev. Lett.* **93** (2004) 150502.
- T. M. Stace, S. D. Barrett, H. -S. Goan, and G. J. Milburn, "Parity measurement of one- and two-electron double well systems", *Phys. Rev. B* **70** (2004) 205342.
- M. Cavallini, J. Gómez, M. Massi, C. Albonetti, D. Ruiz-Molina, C. Rovira, J. Veciana and F. Biscarini, "Magnetic Information Storage on Polymers using Single Molecule Magnets", *Angew. Chem. Int. Ed.* (2004, in press).

- J. Gómez, D. Luneau, K. Wurst, J. Veciana, D. Ruiz-Molina, Ph. Gerbier, "Sterically hindered carboxylates: an investigation of inter-ligand repulsion effects on the synthesis of single-molecule magnets", *New J. Chem.* (2005, in press).
- A. Hernández-Mínguez, M. Jordi, R. Amigó, A. García-Santiago, J. M. Hernandez, J. Tejada, "Low temperature microwave emission from molecular clusters", *Europhys. Lett.* (2005, in press).
- J. Gómez-Segura, M. Cavallini, M. A. Loi, E. Da Como, G. Ruani, M. Massi, M. Muccini, D. Ruiz-Molina, J. Veciana, F. Biscarini, "Self-organised patterns of molecules in a microporous polymer matrix by the breath figures method", *Adv. Mater.* (submitted).
- J. Gómez, D. Ruiz-Molina, J. Veciana, "Concentration and solvent effects on the reorganization of Mn12 single-molecule magnets on the surface of polycarbonate thin-films", *Adv. Funct. Mater.* (submitted).
- G. D. Hutchinson, G. J. Milburn, T. P. Spiller, C. A. Holmes, T. M. Stace, S. D. Barrett, D. G. Hasko and D. A. Williams, "A model for an irreversible current bias in the superconducting qubit measurement process" (*preprint*).
- T. M. Stace and S. D. Barrett, "Continuous measurement of a charge qubit: inelastic tunnelling and the absence of current oscillations", *cond-mat/0309610* (submitted to *New J. Phys.*).
- Ahsan Nazir, Brendon Lovett, Sean Barrett, John H. Reina, Andrew Briggs, "Anticrossings in Foerster Coupled Quantum Dots", *quant-ph/0309099 (Phys. Rev. B.*, to appear).
- S. D. Barrett and T. M. Stace, "Spin readout in exchange interaction quantum computers", (*Phys. Rev. Lett.*, in review).
- S. D. Barrett, P. Kok, K. Nemoto, R. G. Beausoleil, W. J. Munro, and T. P. Spiller, "A symmetry analyser for non-destructive Bell state detection using EIT", *quant-ph/0408117 (Phys. Rev. Lett.*, in review).
- S. D. Barrett and P. Kok, "Efficient high-fidelity quantum computation using matter qubits and linear optics", *quant-ph/0408040* (*Phys. Rev. Lett.*, in review).
- T. M. Stace and S. D. Barrett, "Continuous Quantum Measurement: Stace and Barrett reply", *cond-mat/0406751 (Phys. Rev. Lett.*, in review).
- T. M. Stace, A. C. Doherty, and S. D. Barrett, "Phonon induced population inversion in driven dot quantum systems", (*Phys. Rev. Lett.*, in review).
- S. D. Barrett and T. M. Stace, Continuous measurement of a microwave-driven solid state qubit", (*Phys. Rev. Lett.*, in review).
- W. J. Munro, K. Nemoto, G. J. Milburn and T. P. Spiller, "Condensed matter qubits coupled to a microwave bus" (*in preparation*).
- G. D. Hutchinson, G. J. Milburn, T. P. Spiller, C. A. Holmes, T. M. Stace, "A model for the measurement process in the charge-phase quantronium qubit" (*in preparation*).

#### b. Conferences and invited talks

- Applied Superconductivity Conference (ASC 2002) Texas, USA, 4-9 August 2002; to be published in IEEE Trans. Appl. Supercond. (2003). (poster)
- Oral presentation by Prof. J. Veciana at the *Molecular Nanosystems: From Single Molecules to Supramolecular Assemblies*, Ascona (Switzerland), April 2002.
- Poster by J. Gómez at the *Euresco Conference*, Granada (Spain), June 2002
- Oral presentation by Prof. Jaume Veciana at the *International Conference on Science and Technology of Synthetic Metals*, Shang Hai (China), July 2002.
- Poster by Prof. J. Veciana at *Trends in Nanotechnology (TNT 2002)*, Santiago de Compostela (Spain), September 2002.
- Oral presentation by Dra. C. Rovira at the *International Conference on Molecular Magnetism*, Valencia (Spain), October 2002.
- J. Tejada, R. Amigó, J. M. Hernandez, A. García-Santiago, "Microwave experiments in molecular magnets", proceedings of the *Centenario de las Reales Sociedades Españolas de Física y Química, XXIX Reunión Bienal de Física*, 7-11 July 2003, Madrid (Spain) (poster).
- R. Amigó, J. Tejada, E. M. Chudnovsky, J. M. Hernandez, A. García-Santiago, "Quantum dynamics of crystals of molecular magnets inside microwave resonators", *ICM 2003 International Conference on Magnetism*, 27 July 1 August 2003, Rome (Italy), *Journal of Magnetism and Magnetic Materials* 272-276 (2004) 1106-1108 (poster).
- F. Torres, J. M. Hernandez, A. García-Santiago, J. Tejada, E. Molins, "Experimental evidence of the dependence of spin tunneling on the concentration of dislocations in Mn12 crystals", *ICM 2003 International Conference on Magnetism*, 27 July 1 August 2003, Rome (Italy), *Journal of Magnetism and Magnetic Materials* 272-276 (2004) 1111-1113 (poster).
- J. Tejada, R. Amigó, J. M. Hernandez, A. García-Santiago, "Resonant experiments in magnetism: superradiance and magnetic spectroscopy", *ICM 2003 International Conference on Magnetism*, 27 July 1 August 2003, Rome (Italy), *Journal of Magnetism and Magnetic Materials* 272-276 (2004) 2131-2135 (invited talk).
- J. Tejada, "Microwave experiments in molecular magnets", February 2003, Katholieke Universiteit, Leuven (Belgium) (invited talk).
- L. Hao, J. C. Macfarlane, C. Gardiner, P. Josephs-Franks and J. C. Gallop, "Inductive Superconducting Transition-edge for Photon and macromolecule detection", *Condensed Matter and Materials Physics Conference (CMMP03)*, 6-9 April 2003, Queen's University of Belfast, UK.
- L. Hao, "SQUIDs: Towards the nano-scale", 23 July 2003, invited talk at CSIRO, Lindfield, Sydney, Australia.
- L. Hao, J. C. Macfarlane, P. Josephs-Franks and J. C. Gallop, "Nanoscale Inductive Superconducting Transition-edge Photon and Particle Detector", *Nanoelectronics*, Lancaster University, Lancaster, UK, January, 2003 (poster).
- L Hao, J C Gallop, C. H. Gardiner, P. Josephs-Franks, J C Macfarlane, S K H Lam and C Foley, "Inductive superconducting transition-edge detector for single-photon

- and macro-molecule detection", 9th International Superconducting Electronics Conference 2003 (ISEC2003), 7-11 July 2003, Sydney, Australia (talk).
- P. Josephs-Franks, L Hao, A Tzalenchuk, J Davies, O Kazakova, J C Gallop, L Brown and J C Macfarlane, "Measurement of the spatial sensitivity of miniature SQUIDs using magnetic-tipped STM", 9th International Superconducting Electronics Conference 2003 (ISEC2003), 7-11 July 2003, Sydney, Australia (talk).
- A Tzalenchuk, "Feasibility of ultrasmall Josephson junctions for all-HTS phase qubits", *IOP Superconductivity Annual Conference*, Cambridge, UK, January 2003 (invited talk).
- A Tzalenchuk, "Dynamics and measurement issues of ultrasmall HTS Josephson junctions with unconventional current-phase relation", *Symposium on Quantum Measurements and Metrology*, Gothenburg, Sweden, September 2003 (invited talk).
- A Tzalenchuk, "Fabrication, dynamics, and possible qubit applications of ultrasmall HTS Josephson junctions with unconventional current-phase relation", *International Superconductivity Conference ISS'03*, Tsukuba, Japan, October 2003 (invited talk).
- 12th International Conference on Scanning Tunneling Microscopy/Spectroscopy and related techniques (STM03), 21-25 July 2003, University of Technology, Eindhoven (Netherlands)
- Theoretical Trends in Low Dimensional Magnetism (LDM 2003), 23-25 July 2003, Firenze, Italy.
- N. Ventosa, "Producción de micro- y nano-partículas orgánicas utilizando CO2 comprimido", 1ª Reunión Nacional de la Red Flucomp, Madrid (Spain) 12-14 November 2003.
- N. Ventosa, S. Sala, J. Veciana, "DELOS® Process: Crystallization of Pure Polymorphic Phases from CO<sub>2</sub>-Expanded Solutions", *6th International Symposium on Supercritical Fluids*, Versailles (France), 28-30 April 2003.
- M. Cavallini, M. Massi, F. Biscarini, J. Gómez, D. Ruiz-Molina, C. Rovira, J. Veciana, "Nanopatterning of molecular magnets by stamp-assisted self-organization", *E-MRS Spring Meeting*, Strasbourg (France), 9-13 June 2003.
- M. Cavallini, J. Gómez, M. Massi, F. Biscarini, D. Ruiz-Molina, C. Rovira, J. Veciana, "Deposition and nanopatterning of Mn<sub>12</sub> single-molecule magnets on surfaces with size and positional control at multiple length scales", *Trends in Nanotechnology*, Salamanca (Spain), 15-19 September 2003
- Daniel Ruiz-Molina, "Isolated single-molecule magnets on the surface of a polymeric thin-film", Kinki University, Department of Chemistry, Osaka (Japan), 9 September 2003
- Daniel Ruiz-Molina, "Isolated single-molecule magnets on the surface of a polymeric thin-film", Osaka University, Department of Chemistry, Osaka (Japan), 12 September 2003. J. Veciana, "Functional Supramolecular Nano-Architectures", Max Planck Institut, Stuttgart (Germany), 1 December 2003. 4th Quantum Information Processing and Communication (QIPC) Workshop organized by Quiprocone (www.quiprocone.org), Oxford (UK), 13-17 July 2003 (T. P. Spiller was the organizer).

- Few Qubit Applications, Budmerice (Slovakia), 11-14 December 2003 (two invited talks).
- *1st QuICT (Quantum Interference, Correlation and Technology) Network meeting*, Abingdon (UK), 11-13 July 2003. (Talk: "Quantum measurement in solid state quantum computers").
- 4th QIPC Workshop, Oxford (UK), 13-17 July 2003 (1 poster: "Quantum measurement in solid state quantum computers").
- Solid State Quantum Information Processing (SSQIP) Conference, Amsterdam (The Netherlands), 15-18 December 2003 (1 talk, 3 posters).
- J. Tejada, A. Hernández, M. Jordi, R. Amigó, J. M.Hernández, "Emission of electromagnetic radiation from molecular magnets", *International Conference on the Low Energy Electrodynamics in Solids (LEES04)*, Kloster Banz (Germany), 18-23 July 2004. (Oral)
- A. Hernández-Mínguez, M. Jordi, R. Amigó, A. García-Santiago, J. M. Hernández and J. Tejada, "Collective microwave emision from molecular clusters at low temperatures", *International Conference on the Low Energy Electrodynamics in Solids (LEES04)*, Kloster Banz (Germany), 18-23 July 2004. (Poster)
- J. Tejada, A. Hernández, R. Amigó, M. Jordi, J. M.Hernández, "Quantum Tunneling in Molecular Magnets", *XI International Summer School "Nicolás Cabrera", Frontiers in Science and Technology: Magnetic Nanostructures*, Miraflores de la Sierra, Madrid (Spain), 13-17 September 2004. (invited talk)
- A. Hernández-Mínguez, M. Jordi, R. Amigó, A. García-Santiago, J. M. Hernández and J. Tejada, "Collective microwave emision from molecular clusters at low temperatures", *International Conference on Molecule-Based Magnets (ICMM 2004)*, Tsukuba, Japan, 4-8 October 2004. (Poster)
- M. Jordi, J. Tejada, R. Amigó, J. Vanacken, S. Stroobants, M. Malfait, V. V. Moshchalkov, E.M. Chudnovsky and D.A. Garanin, "Magnetization reversal in molecular nanomagnets under pulsed magnetic fields", *International Conference on Molecule-Based Magnets (ICMM 2004)*, Tsukuba, Japan, 4-8 October 2004. (Poster)
- Javier Tejada, "Molecular Magnets and microwaves", City University of New York, 17 March 2004.
- Javier Tejada, "Molecular magnets and superradiance", New York University, 18 March 2004.
- Javier Tejada, "Microwave radiation from molecular clusters", *March Meeting 2004*, American Physical Society, Montreal (Canada).
- Javier Tejada, "The route from single domain particles to molecular clusters", University of Stuttgart, 16 July 2004.
- L. Hao, J. C. MacFarlane, J. C. Gallop, and S. Lam, "Inductive sensor based on nano-scale SQUIDs", *Applied Superconductivity Conference 2004 ASC'04*, Jacksonville, Florida, USA, October 3-8 2004 (to be published).
- Condensed Matter and Materials Physics Conference (CMMP04), 4-7 April 2004, University of Warwick, UK.

- Conference on Precision Electromagnetic Measurements (CPEM 2004), 27 June 2 July 2004, London (UK).
- 4th International Symposium on Metallic Multilayers (MML04), 7-11 June 2004, Boulder, Colorado (USA).
- D. Ruiz-Molina, J. Gómez-Segura and J. Veciana, "Multiple Length Scale Patterning of Single-Molecule Magnets", *1st NanoSpain Workshop*, San Sebastian (Spain), 10-12 March 2004. (Oral)
- D. Ruiz-Molina, J. Gómez-Segura and J. Veciana, "Multiple Length Scale Patterning of Single-Molecule Magnets", *III Reunión Nacional de Física del Estado Sólido*, San Sebastian (Spain), 2-4 June 2004. (Oral)
- J. Gómez-Segura, D. Ruiz-Molina and J. Veciana, "Nanopatterning of Single-Molecule Magnets on a surface", *IV Congreso Español de Fuerzas y Túnel*, Vic (Spain), 21-24 September 2004. (Oral)
- J. Gómez-Segura, D. Ruiz-Molina, C. Rovira and J. Veciana, "Multiple Length Scale Patterning of Single-Molecule Magnets", *Quantum Effects on Molecular Nanomagnets (QuEMolNa Kick-off meeting)*, Valencia (Spain), 28-29 May 2004. (Oral)
- J. Gómez-Segura, D. Ruiz-Molina, C. Rovira and J. Veciana, "Nanopatterning of Single-Molecule Magnets on a surface", 7th International Conference on Nanostructured Materials, Wiesbaden (Germany), 20-24 June 2004. (Poster)
- J. Gómez-Segura, D. Ruiz-Molina, C. Rovira and J. Veciana, "Nanopatterning of Single-Molecule Magnets on a surface", *International Conference on Molecular Magnets*, Tsukuba (Japan), 04-08 September 2004. (Oral)
- J. Veciana, "Functional Molecular Nano-Objects", *Jornades Franco-Catalanes de Nanociències i Nanotecnologia*, Barcelona (Spain) 5-6 July 2004.
- J. Veciana, "Nanopatterning of Mn<sub>12</sub> single-molecule magnets on surfaces. Towards magnetic information storage", *Workshop on "Perspectives on Single-Molecule and Single-Chain Magnets" CREST, JST*; Tsukuba (Japan), October 4th, 2004.
- T. M. Stace and S. D. Barrett, "Charge qubit Rabi oscillations in point contact detectors", *Quantum Dots Conference (QD2004)*, 10-13 May 2004, Banff, Alberta (Canada). (Poster)
- S. D. Barrett and T. M. Stace, "Continuous measurement of a microwave-driven solid state qubit", *X International Conference on Quantum Optics (ICQO2004)*, 30 May 3 June 2004, Minsk (Belarus). (Invited talk)
- S. D. Barrett and T. M. Stace, "Continuous measurement of a microwave-driven solid state qubit: an all-electrical analogue of resonance-fluorescence *Seventh International Conference on Quantum Communication, Mesaurement and Computing* (QCMC2004), 25-29 July 2004, Glasgow (UK). (Talk)
- S. D. Barrett and T. M. Stace, "Techniques for spin measurement in exchange interaction quantum computers", *Seventh International Conference on Quantum Communication, Mesaurement and Computing (QCMC2004)*, 25-29 July 2004, Glasgow (UK). (Poster)

- T. M. Stace and S. D. Barrett, "The role of inelastic tunnelling in a continuous quantum measurement of a solid state qubit", *Seventh International Conference on Quantum Communication, Mesaurement and Computing (QCMC2004)*, 25-29 July 2004, Glasgow (UK). (Poster)
- B. W. Lovett, A. Nazir, S. D. Barrett, T. P. Spiller, and G. A. D. Briggs, "Selective Spin Coupling using a Single Exciton", *Mesoscopic Quantum Coherence and Computing*, 7-10 June 2004, Napoli (Italy). (Talk)
- S. D. Barrett and P. Kok, "Efficient high-fidelity quantum computation using matter qubits and linear optics", *QISW04* Cambridge (UK) (2004). (Poster)
- B. W. Lovett, A. Nazir, S. D. Barrett, T. P. Spiller, and G. A. D. Briggs, "Selective Spin Coupling using a Single Exciton", QISW04 Cambridge (UK) (2004). (Poster)
- W. J. Munro, K. Nemoto, T. P. Spiller, S. D. Barrett, R. G. Beausoleil, and P. Kok, "Linear and nonlinear quantum optical information processing". (Invited talk)

In addition to these international events listed above, HPLB members presented results at a number of smaller UK meetings (*UK EPSRC Quantum Circuits Network Meetings*, *Oxford Quantum Logic Gate Club Meetings*), and gave a number of seminars at various UK Universities and other institutions internationally, including the University of Queensland, the Centre for Quantum Computer Technology (University of New South Wales), and Harvard University.

# 6. Potential impact of project results

The different workpackages have delivered very interesting results along the three years, all of them innovative and state-of-the-art within the field, that have impact on many areas as described below.

# WP1. MicroSQUID and Hall probe development

<u>Verification of SQUID sensitivity with loop area</u>. It was shown that the sensitivity of commercially produced, lithographically patterned SQUIDs, scales as expected by theory down to the micron range. This impacts on the sensitivity obtainable from such devices which can be used as extremely sensitive detectors for a number of potential applications.

<u>Development of sub-micron loop SQUID devices</u>. It was shown that it is possible to produce, using e-beam lithography and other techniques, SQUID devices with loop areas well below the micron range. This impacts on the development of such devices as ultra miniature detectors of radiation as well as their suitability to magnetic qubit detectors. The system is scalable and thus paves the way for arrays of such devices to be produced impacting on several socio-economic areas.

Fabrication technology for superconducting circuits and devices. The technology developed for this project has a foreseable impact on the know-how portfolio of the CSIC-IMM unit. Gained experience with this type of devices is extremely valuable to approach new developments in the next future. In particular, CSIC-IMM plans to take part in a national and European initiative to develop an X-Ray spectrometer for the European Space Agency based on superconducting bolometers also requiring quantum limited detection at very low temperatures. This would have been impossible without the training and experience gained through the project. On the other hand, electron-beam lithography, which was optimized in order to obtain large area contacts in combination with the smallest features such as nano-constrictions, has large potential for the fabrication of many other devices. Training achieved through this pioneer project will have an impact on the operation of two other EBL systems in the Universitat Autònoma de Barcelona and the Universitat de Zaragoza.

### WP2. Qubit preparation, identification and characterisation

<u>Preparation of clusters</u>. The work performed involved mostly the synthesis and characterization of new  $Mn_{12}$  clusters with interesting optically-active ligands to find synergistic effects. Two complementary techniques, magnetic susceptibility and magnetic circular dichroism (MCD), were used and it was shown, for the first time, that the magnetic and optical responses are essentially identical. The development of new methods for measurement magnetic and spin properties of isolated molecules is important when the detection limits of traditional SQUID instruments are reached.

<u>Obtaining of nanoparticles</u>. A new crystallization technique based on the use of supercritical fluids was set up. Although their use is well-known for instance, for the controlled crystallization and delivery of pharmaceutical drugs, this was the first time that this technique was used to obtain nanocrystals of molecular magnets. This fact is not only important because the nanocrystals obtained in this way exhibit a small dispersion range, but also because their size and shape can be controlled at will, influencing their properties when compared with the materials obtained by traditional crystallization techniques.

## WP3. Qubit deposition and purification

<u>Surface deposition</u>. A simple process that is able to fabricate patterns of magnetic bits both on a polymer or on different surfaces (HOPG, SiO<sub>2</sub>, etc.) was developed by CSIC-ICMAB. It was demonstrated that this system can be effectively used as a permanent not only for quantum computation but also for information storage medium with magnetic readout. This technique may lead to the fabrication of magnetic Mn<sub>12</sub> domains with features a few tens of nanometers in size, which corresponds to a few molecular diameters (end-to-end distance is 4 nm). Finally, the patterning and addressing of molecular materials across multiple length scales is one of the most important issues in nanotechnology. Extension of this work could lead to the development of next-generation storage media based on a variety of molecular responses, for example: charge storage; conformational, orientational, or positional shifting; phase transitions or reorganization; and optical anisotropy changes and fluorescence switching.

#### WP4. Microwave and RF measurements

Due to the work performed on the search of the magnetic qubits we came to the idea that superradiance emission could interfere with the operation of these qubits as logic quantum gates as it would determine entirely the time of their operation. Our findings of the dependence of the fast reversal of the magnetic moment on the sweep rate of the applied field when we were looking at the fast magnetic response of the magnetic qubits was also interpreted as the signal that superradiance could be present when operating with a large number of magnetic qubits.

We are fully convinced that our results about the electromagnetic radiation emission by molecular magnets open a new field of research in the intersection of quantum mechanics, magnetim and microwaves. Concerning the results about the fast magnetization reversal, the novelty lies on our suggestion that we are mostly dealing with a new quantum relaxation mechanism which involves the coherent rotation of a macroscopic number of molecules. This result enters then the category of a new macroscopic quantum phenomena.

It is also important to remark that both results have technological implications as we are suggesting that the molecular magnets are natural sources of coherent electromagnetic radiation in the frequency range between GHz and THz. In the scope of quantum computation, we should now have in mind that their operation time is mostly limited by the emission of radiation. In case of making possible to have molecular magnets acting as strong sources of electromagnetic radiation, we may anticipate that these materials

will have a very big impact in areas such as medicine, security and telecommunications. With our work it is also clear that Europe is leading this new research field. During the three years of duration of the project, we were invited to give several talks in both international meetings and universities and reseach centers.

## WP5. Development of measurement tools for quantum nano-magnets

<u>Magnetic imaging of SQUIDs using magnetic tipped STM</u>. A novel method of using a SQUID to detect its own sensitivity using a magnetic tipped STM system was developed. This impacts in several areas as it shows the ability to use a magnetic tipped STM to achieve magnetic information on a very minute length scale and also shows the ability to combine two quantum detection systems, an STM and a SQUID in one instrument allowing a combination of topography and magnetic information to be obtained simultaneously. This has potential to impact on several technologically significant areas from magnetic storage to quantum computation.

<u>Imaging and interaction of  $Mn_{12}$  with AFM/MFM</u>. We achieved the imaging and manipulation of  $Mn_{12}$  using MFM and AFM systems. This has impact on several areas where the precise positioning of nanoscale objects is required. This impacts on a wide variety of areas and is a vital tool if nanotechnology is to be developed in to a disruptive technology that it predicted to do so.

<u>Development of SAW interdigitated transducers on LiNbO</u><sub>2</sub>. Technology for surface acoustical waves (SAW) transducers will impact also on future measurement techniques and applications in the biosensor field.

### WP6. Exploration of the fundamentals of quantum nano-magnets

<u>Testing of SQUID devices down to 50 mK using low noise techniques</u>. Using an adiabatic demagnetisation refrigerator (ADR) system we were able to cool and operate micro-SQUIDs down to 50 mK. This has impact on other potential applications that will need quantum limited detection methods operating at very low temperatures. The use of nanoscale SQUIDs at such low temperatures, for example as bolometers, could also impact on a wider technology area.

<u>Use of high- $T_c$ </u> <u>Josephson junctions as potential THz sources/detectors</u>. THz sources and detectors are of interest to a wide variety of areas that could impact on the quality of life. Using high temperature superconductors as both sources and detectors could have potential impact to a wide variety of applications.

### WP7. Theory and simulations

The theory and modelling effort delivered results in seven general areas during the course of the project. These have impact for experiments on magnetic qubits, but also wider impact for solid state QIP, as described below:

<u>Study of the continuous measurement of solid state qubits</u>. This yielded understanding of the fundamental decoherence effects induced by various measurement schemes into the qubits they are measuring was reached. This is relevant for magnetic qubits probed by SQUIDs, but has rather wider impact in being applicable to other solid state qubit systems.

<u>Understanding of the operation of a DC SQUID</u> or large current-biased Josephson junction <u>operating as a qubit measurement system</u>, in particular the knock-on effects to the qubit of the intrinsic irreversibility of a current fed to the measurement apparatus. This has impact for the use of such SQUID systems in any quantum measurement scenario.

<u>Understanding of the behaviour of a microwave-driven solid state qubit subject to decoherence</u> – the effect of the Rabi oscillation signal on the measurement apparatus. This has impact for any solid state system driven with an external classical oscillatory field. In particular, various useful parameters concerning the qubit and its environment can be extracted from this measurement scenario.

<u>Study of the readout process</u> (continuous observation and single shot projection) <u>for matter qubits</u>. Conventional approaches to QIP assume the existence of single shot projective readout for qubits, so this is the ultimate aim for any qubit system.

<u>Novel proposals for hybrid matter/optical QIP</u>. In the short term it is important to simply get solid state qubits working. However, taking a longer term perspective and thinking about actual QIP devices and technology, it is likely that communication will come into play. Hybrid matter/optical QIP systems are likely to play a very important role here.

<u>Study of the probing of magnetic qubits using external thermal noise</u>. This is a useful investigative technique for plotting out magnetic susceptibilities of qubits. The groundwork was done many years ago by one of the participants for SQUID systems – this can be readily adapted for nanomagnets.

<u>New approaches to detection and quantum gates based on the interactions</u> between magnetic qubits and a common microwave bus mode. This work can be applied to any solid state qubits that couple to microwaves. Further ideas continue to emerge.

## 7. Future outlook

In the chapter of preparation of molecular magnets, the patterning process can be scaled down to smaller length scales and higher densities by improving the resolution of the master, or by patterning the solute/polymer mixture by using a higher-resolution fabrication technique, for example, nanoimprint lithography. This technique may lead to the fabrication of magnetic  $Mn_{12}$  domains with features a few tens of nanometers in size, which corresponds to a few molecular diameters (end-to-end distance is 4 nm). Another important venue would be to examine the feasibility of these clusters not only as qubits but also as information units for high-density storage devices.

The main work concerning measuring devices was the development and testing of micro-SQUIDs. These ranged from commercially produced ones, with loops of the order of a few microns, down to state-of-the-art niobium devices from CSIRO in Australia through a collaboration with NPL. It was shown that the commercially produced devices scale their spin sensitivity with their area as expected from theory, and indeed these devices shown the best spin sensitivity of the order of a few tens of spins per unit bandwidth. The production of even small SQUIDs through e-beam lithography and FIB can enable devices with loops in the nanometre range to be produced. However, with such small dimensions it is clear that the quality of the superconducting films has to be of the highest quality otherwise the superconductivity is soon lost. We shown that it is possible to produce SQUIDs with loops of the order of a few nanometers that should be able to detect close to the single spin limit required for qubit readout. However, more work is needed to carefully taylor the properties of these devices so that this sensitivity is realised. For example, we found that self-heating to the geometric structure of the weak links can be a problem so careful design is also required. Also a gold layer on top of the niobium can act as a shunt resistance to prevent hysteresis at low temperatures. Work is needed to investigate this futher and to taylor specific devices with well defined operating temperatures and dimensions so that all of their properties can be optimised to enable qubit readout.

According to CSIC-IMM, the use of micro-SQUID devices for the detection of single spin transitions seems to have reached its practical limits and other type of quantum limited detection approach needs to be investigated in the next future. This unit foresees to work on the field of single particle magnetic resonance detection with nanomechanical transducer devices, that it is estimated very promising in combination with CSIC-IMM unique technical skills in the III-V semiconductors growth and processing. The experimental approach opened by UB on magnetic measurements in the microwave range it is also a very promising field and CSIC-IMM will try to collaborate by supplying SAW type devices, waveguides and bolometers.

NPL will further investigated the means of  $Mn_{12}$  deposition within SQUID loops. It appears that AFM manipulation may be a possible mechanism but it is a somewhat slow process, to move much smaller clusters it needs to have investigated the possibility of using very sharp AFM tips, perhaps with carbon nanotubes attached to the tips. We consider it unlikely that the SQUIDs could be constructed around a pre-patterned array of  $Mn_{12}$ , thus some manipulation means is needed or a means of maximising the serendipity of a chance deposition within a SQUID loop.

 $Mn_{12}$  certainly has interesting properties and is a very good candidate for the study of nanomagnetism and quantum effect. We believe that the study of the physics involved should be pursued. The use of high temperature superconducting Josephson junctions as a means to detect radiation up to several THz may be an important means of carrying out these investigations.

We are pretty optimistic about the fact that with the work performed in the field of microwave radiation along the last three years we have opened a new research area which may be of large importance for both science and technology. In the field of basic science, it is worth to mention that variants of some of the results of our experiments may be catalogued as new manifestations of the Bose-Einstein condensate. In this scope of basic science it is important also to mention that with more experiments it could be possible to go deeper in the understanding of the spin–phonon interaction, opening the door for the research of the interaction between spins and other collective excitations, like for example surface acoustic waves and ferroelectric excitations. The interaction of electromagnetic radiation with the spin system will also be much better studied using our results as a basis for performing new experiments.

In the case of applications it is important to mention that we still have to validate (1) the existence of coherent radiation at room temperature, (2) if the maximum power of the emitted radiation is of the order or larger than those obtained form the present used sources, and (3) the possibility to use this radiation in medicine and telecommunications and security because of its high imaging resolution. In the case of quantum computing, it seems that it will be worth working deeper, as it is clear now that we are mostly and only limited by the decoherence time introduced by the superradiance emission. The importance of our work for the future development in the field of quantum computing can be then summarised saying that it is important to go further in the understanding of the interaction of the spin states with the phonon bath and nuclear spins as well as with the topological arrangements of single qubits.

Much of the theoretical effort over the last two years focussed on generic questions regarding the implementation of solid state quantum computation. As nanomagnet and SQUID detector technology improves such that individual systems can be coherently manipulated and observed, it will be appropriate to tailor this general work to the specific case of nanomagnets and SQUID detectors.

The generic work on microwave driven qubits could be applied to the driven single nanomagnet-SQUID system. This will involve considering the specifics of the nanomagnet Hamiltonian, the effect of the decohering environment of the nanomagnet, and the coupling to the detector. Predictions of the SQUID output would allow comparison with experiment, and allow useful spectroscopic information, such as single nanomagnet coherence times, to be extracted from experimental data. Similarly, many of the ideas for qubit readout can also be applied in the context of nanomagnetic qubits. For example, the noise probing technique can be applied directly to the detection of magnetic qubit susceptibilities. It is a straightforward task to model directly measured noise profiles, once details of the integration window and couplings are known.

Another avenue of future enquiry would be to examine the possibility of implementing a hybrid quantum computing scheme using nanomagnets. On the face of it, the hybrid matter-optical schemes described above would seem to be ruled out, on account of the absence of an optical transition in the nanomagnet systems under consideration. However, it should be possible to couple individual nanomagnets to superconducting circuits, resonant to a microwave transition in the nanomagnet. This opens up the application of many new techniques currently being developed in quantum optics to the arena of solid state qubits such as nanomagnets. One could then investigate whether the hybrid matter-optical schemes described above can be implemented in a hybrid nanomagnet-microwave circuit system, e.g. to entangle distant nanomagnet qubits, or to use microwaves as a measurement or conditioning tool for nanomagnets. All this is an extremely interesting research direction that should generate further important new results over the next few years.

# 8. Appendix 6. Project's achievements fiche

Questions about project's outcomes	Number	Comments
1. Scientific a	nd technolog	gical achievements of the project (and why are they so ?)
Question 1.1.  Which is the 'Breakthrough' or 'real' innovation	N/A	Brief description:
achieved in the considered period	1771	A new way has been developed to deposit and/or nanopattern aggregates or isolated Mn <sub>12</sub> molecules that can be use a qubits.
		Successful fabrication of 200 nm loop diameter Nb single layer SQUIDs in collaboration with CSIRO Australia.
		Use of high temperature superconducting Josephson junctions as THz detectors and sources.
		• Improvement of the patterning of Mn <sub>12</sub> SMMs with size and distance control on a lithographically patterned micro-SQUID.
		Preparation of a series of chiral dodecamanganese clusters that behave as single-molecule magnets along with optical activity.
		The GAS crystallization process, based on supercritical fluids, has been successfully used for the controlled preparation of nanocrystals of Mn <sub>12</sub> .
		<ul> <li>Development of a new methodology by which Mn<sub>12</sub> clusters are patterned on a surface with controlled size and distributions by molding a dispersion of the cluster in a polymer matrix onto a structured master and posterior treatment with solvent.</li> </ul>
		• Single crystals of a deuterated and pure sample of the well-known Mn <sub>12</sub> Ac were obtained.
		• A new method of SQUID self-portraiture has been developed in which a magnetic scanned probe tip is scanned over a micro-SQUID whilst simultaneously recording the SQUID response. This enables the SQUID spatial sensitivity to be mapped thus allowing a determination of the ideal spin location position within the SQUID device.
		A new method that determines the quality factor of microwave resonant cavities, and can be used as well as a new magnetic spectrometer, has been developed. The idea is to use the magnetic moment of molecular magnets inside cavities to determine the power and the spectrum of the microwave arriving the cavity.

	A new idea of getting microwave superradiance from the rapid demagnetization of molecular magnets has been presented.
	<ul> <li>Determination of the influence of sample geometry, temperature and magnetic field on the radiation emitted by molecular magnets.</li> </ul>
	Distinguishing between black body radiation and radiation emitted by molecular magnets.
	Resolving the variation of magnetization and the detection of radiation in time.
	• Detection of superradiance emission by Mn <sub>12</sub> molecular clusters.
	<ul> <li>Understanding of the fundamental decoherence effects induced by various measurement schemes into the qubits they are measuring has been reached.</li> </ul>
	Understanding of the operation of a DC SQUID or large current-biased Josephson junction operating as a qubit measurement system
	• Understanding of the behaviour of a microwave-driven solid state qubit subject to decoherence – the effect of the Rabi oscillation signal on the measurement apparatus.
	• Study of the readout process (continuous observation and single shot projection) for matter qubits.
	Novel proposals for hybrid matter / optical QIP.
	Study of the probing of magnetic qubits using external thermal noise.
	New approaches to detection and quantum gates based on the interactions between magnetic qubits and a common microwave bus mode.
2 1	
	ence and Technology: Scientific Publications in scientific magazines
Question 2.1.	<u>UB</u> :
Scientific or technical publications on reviewed journals and conferences	<ul> <li>R. Amigó, E. del Barco, Ll. Casas, E. Molins, J. Tejada, I. B. Rutel, B. Mommouton, N. Dalal, J. Brooks, "Quadratic Transverse Anisotropy Term due to Dislocations in Mn<sub>12</sub> Acetate Crystals Directly Observed by EPR Spectroscopy", <i>Physical Review B</i> 65 (2002) 172403.</li> </ul>
	• J. M. Hernandez, F. Torres, J. Tejada, E. Molins, "Crystal Defects and Spin Tunneling in Single Crystals of Mn <sub>12</sub> Clusters", <i>Physical Review B</i> <b>66</b> (2002) 161407(R).

- R. Amigó, J. M. Hernandez, A. García-Santiago, J. Tejada, "High-resolution detection of resonant frequencies of microwave resonators via magnetic measurements", *Applied Physics Letters* **82** (2003) 4528-4530.
- R. Amigó, J. M. Hernandez, A. García-Santiago, J. Tejada, "Microwave absorption and magnetization tunnelling in Mn<sub>12</sub>-acetate molecular clusters", *Physical Review B* **67** (2003) 220402(R).
- R. Amigó, J. M. Hernandez, A. García-Santiago, J. Tejada, "Magnetic detection of millimeter waves", *Europhysics Letters* **64** (2003) 158-163.
- J. Tejada, R. Amigó, J. M. Hernandez, E. M. Chudnovsky, "Quantum dynamics of crystals of molecular nanomagnets inside a resonant cavity", *Physical Review B* 68 (2003) 014431.
- R. Amigó, J. Tejada, E. M. Chudnovsky, J. M. Hernandez, A. García-Santiago, "Quantum dynamics of crystals of molecular magnets inside microwave resonators", *ICM 2003 International Conference on Magnetism*, 27 July 1 August 2003, Rome (Italy), *Journal of Magnetism and Magnetic Materials* 272-276 (2004) 1106-1108.
- F. Torres, J. M. Hernandez, A. García-Santiago, J. Tejada, E. Molins, "Experimental evidence of the dependence of spin tunneling on the concentration of dislocations in Mn<sub>12</sub> crystals", *ICM 2003 International Conference on Magnetism*, 27 July 1 August 2003, Rome (Italy), *Journal of Magnetism and Magnetic Materials* 272-276 (2004) 1111-1113.
- M. Jordi, A. Hernandez-Mínguez, J. M. Hernandez, J. Tejada, S. Stroobants, J. Vanacken and V. V. Moshchalkov, "Scaling of the susceptibility vs. magnetic-field sweep rate in molecular magnet", *Europhys. Lett.* 68 (2004) 888.
- J. Vanacken, S. Stroobants, M. Malfait, V. V. Moshchalkov, M. Jordi, J. Tejada, R. Amigó, E. M. Chudnovsky, D. Garanin, "Pulsed field studies of the magnetization reversal in molecular nanomagnets", *Phys. Rev B* 70 (2004) 220401.
- A. Hernández-Mínguez, M. Jordi, R. Amigó, A. García-Santiago, J. M. Hernandez, J. Tejada, "Low temperature microwave emission from molecular clusters", *Europhys. Lett.* (2005, in press).

#### CSIC:

P. Gerbier, D. Ruiz-Molina, J. Gómez, K. Wurst, J. Veciana, "Examining Thermolysis Reactions of Nanoscopic Mn<sub>12</sub> Single Molecule Magnets", *Polyhedron* 22 (2003) 1951.

- M. Cavallini, F. Biscarini, J. Gómez, D. Ruiz-Molina, J. Veciana, "Multiple length scale patterning of single-molecule magnets", *Nano Letters* **3** (2003) 1527.
- M. Cavallini, J. Gómez, M. Massi, C. Albonetti, D. Ruiz-Molina, C. Rovira, J. Veciana and F. Biscarini, "Magnetic Information Storage on Polymers using Single Molecule Magnets", *Angew. Chem. Int. Ed.* (2004, in press).
- J. Gómez, D. Luneau, K. Wurst, J. Veciana, D. Ruiz-Molina, Ph. Gerbier, "Sterically hindered carboxylates: an investigation of inter-ligand repulsion effects on the synthesis of single-molecule magnets", *New J. Chem.* (2005, in press).
- J. Gómez-Segura, M. Cavallini, M. A. Loi, E. Da Como, G. Ruani, M. Massi, M. Muccini, D. Ruiz-Molina, J. Veciana, F. Biscarini, "Self-organised patterns of molecules in a microporous polymer matrix by the breath figures method", *Adv. Mater.* (submitted).
- J. Gómez, D. Ruiz-Molina, J. Veciana, "Concentration and solvent effects on the reorganization of Mn<sub>12</sub> single-molecule magnets on the surface of polycarbonate thin-films", *Adv. Funct. Mater.* (submitted).

#### CSIC + UB:

- D. Ruiz-Molina, M. Mas-Torrent, A. I. Balana, N. Domingo, J. Tejada, M. T. Martínez, C. Rovira, J. Veciana, "Isolated Single-Molecule Magnets on the Surface of a Polymeric Thin Film", *Adv. Mater.* **15** (2003) 42.
- P. Gerbier, D. Ruiz-Molina, N. Domingo, D. B. Amabilino, J. Vidal-Gancedo, J. Tejada, J. Veciana, "Synthesis and Characterization of a [Mn<sub>12</sub>O<sub>12</sub>(O<sub>2</sub>CR)<sub>16</sub>(H<sub>2</sub>O)<sub>4</sub>] Complex Bearing Paramagnetic Carboxylate Ligands. Use of a Modified Acid Replacement Synthetic Approach", Monatshefte für Chemical Monthly 134 (2003) 265.
- N. Domingo, P. Gerbier, J. Gómez, D. Ruiz-Molina, D. B. Amabilino, J. Tejada, J. Veciana, "Synthesis and characterization of a new chiral nanomagnet", *Polyhedron* 22 (2003) 2355.
- D. Ruiz-Molina, M. Mas-Torrent, A. I. Balana, N. Domingo, J. Tejada, M. T. Martínez, C. Rovira, J. Veciana, "Single-molecule magnets on a polymeric thin film as magnetic quantum bits", SPIE Proceedings 5118 (2003) 594.
- N. Domingo, B. E. Williamson, J. Gómez-Segura, Ph. Gerbier, D. Ruiz-Molina, D.B. Amabilino, J. Veciana, J. Tejada, "Magnetism of Isolated Mn<sub>12</sub> Single-molecule Magnets Detected by Magnetic Circular Dichroism: Observation of Spin Tunneling with a Magneto-optical Technique",

Physical Review B 69 (2004) 052405.

Ph. Gerbier, N. Domingo, J. Gómez, D. Ruiz-Molina, D. B. Amabilino, J. Tejada, B. E. Williamson, J. Veciana, "Chiral, Single-Molecule Nanomagnets: Synthesis, Magnetic Characterization and Natural and Magnetic Circular Dichroism", J. Mater. Chem. 14, 2455 (2004).

#### NPL:

- J. Gallop, P. W. Josephs-Franks, J. Davies, L. Hao, J. Macfarlane, "Miniature dc SQUID devices for the detection of single atomic spin-flips", *Physica C* **368** (2002) 109-113.
- L. Hao, J. C. Gallop, C. Gardiner, P. Josephs-Franks, J. C. Macfarlane, S. K. H. Lam, C. Foley, "Inductive superconducting transition-edge detector for single-photon and macro-molecule detection", *Superconductor Science and Technology* **16** (2003) 1479-1482.
- P. W. Josephs-Franks, L. Hao, A. Tzalenchuk, J. Davies, O. Kazakova, J. C. Gallop, L. Brown, J. C. Macfarlane, "Measurement of the spatial sensitivity of miniature SQUIDs using magnetic-tipped STM", Superconductor Science and Technology 16 (2003) 1570-1574.
- J. C. Gallop, "SQUIDs: some limits to measurement", Superconductor Science and Technology 16 (2003) 1575-1582.
- L. Hao, J. C. Macfarlane, P. W. Josephs-Franks, J. C. Gallop, "Inductive superconducting transition edge photon and particle detector", *IEEE Transactions on Applied superconductivity* **13** (2003) 622-625.

#### HPLB:

- B Schelpe, A Kent, W Munro, T Spiller, "Inferring superposition and entanglement from measurements in a single basis", *Physical Review A* **67** (2003) 052316.
- A. Nazir, B. W. Lovett, S. D. Barrett, T. P. Spiller, and G. A. D. Briggs, "Selective spin coupling through a single exciton", *Phys. Rev. Lett.* **93** (2004) 150502.
- T. M. Stace, S. D. Barrett, H. -S. Goan, and G. J. Milburn, "Parity measurement of one- and two-electron double well systems", *Phys. Rev. B* **70** (2004) 205342.
- Ahsan Nazir, Brendon Lovett, Sean Barrett, John H. Reina, Andrew Briggs, "Anticrossings in Foerster Coupled Quantum Dots". *quant-ph/0309099*, *Physical Review B* (to appear).
- T. M. Stace and S. D. Barrett, "Continuous quantum measurement: inelastic tunnelling suppresses

	1	
		current oscillations" cond-mat/0307088, submitted to Physical Review Letters.
		• T. M. Stace and S. D. Barrett, "Continuous measurement of a charge qubit: inelastic tunnelling and the absence of current oscilations" <i>cond-mat/0309610</i> , submitted to <i>Physical Review B</i> .
		• S. D. Barrett and T. M. Stace, "Spin readout in exchange interaction quantum computers", ( <i>Phys. Rev. Lett.</i> , in review).
		• S. D. Barrett, P. Kok, K. Nemoto, R. G. Beausoleil, W. J. Munro, and T. P. Spiller, "A symmetry analyser for non-destructive Bell state detection using EIT", <i>quant-ph/0408117 (Phys. Rev. Lett.</i> , in review).
		• S. D. Barrett and P. Kok, "Efficient high-fidelity quantum computation using matter qubits and linear optics", <i>quant-ph/0408040 (Phys. Rev. Lett.</i> , in review).
		T. M. Stace and S. D. Barrett, "Continuous Quantum Measurement: Stace and Barrett reply", cond-mat/0406751 (Phys. Rev. Lett., in review).
		• T. M. Stace, A. C. Doherty, and S. D. Barrett, "Phonon induced population inversion in driven dot quantum systems", ( <i>Phys. Rev. Lett.</i> , in review).
		• S. D. Barrett and T. M. Stace, Continuous measurement of a microwave-driven solid state qubit", ( <i>Phys. Rev. Lett.</i> , in review).
		• W. J. Munro, K. Nemoto, G. J. Milburn and T. P. Spiller, "Condensed matter qubits coupled to a microwave bus" ( <i>in preparation</i> ).
		G D Hutchinson, G J Milburn, T P Spiller, C A Holmes and T M Stace, "A model for the measurement process in the charge-phase quantronium qubit", in preparation.
Question 2.2.  Scientific or technical publications on non-reviewed journals and conferences	25	Title and journals/conference and partners involved  UB:
journals and conferences 2		<ul> <li>J. Tejada, R. Amigó, J. M. Hernandez, A. García-Santiago, "Microwave experiments in molecular magnets", proceedings of the <i>Centenario de las Reales Sociedades Españolas de Física y Química, XXIX Reunión Bienal de Física</i>, Madrid (Spain), 7-11 July 2003.</li> </ul>
		J. Tejada, "Microwave experiments in molecular magnets", February 2003, Katholieke Universiteit, Leuven (Belgium).

- J. Tejada, A. Hernández, M. Jordi, R. Amigó, J. M.Hernández, "Emission of electromagnetic radiation from molecular magnets", *International Conference on the Low Energy Electrodynamics in Solids (LEES04)*, Kloster Banz (Germany), 18-23 July 2004.
- A. Hernández-Mínguez, M. Jordi, R. Amigó, A. García-Santiago, J. M. Hernández and J. Tejada, "Collective microwave emision from molecular clusters at low temperatures", *International Conference on the Low Energy Electrodynamics in Solids (LEES04)*, Kloster Banz (Germany), 18-23 July 2004. (Poster)
- J. Tejada, A. Hernández, R. Amigó, M. Jordi, J. M.Hernández, "Quantum Tunneling in Molecular Magnets", XI International Summer School "Nicolás Cabrera", Frontiers in Science and Technology: Magnetic Nanostructures, Miraflores de la Sierra, Madrid (Spain), 13-17 September 2004. (invited talk)
- A. Hernández-Mínguez, M. Jordi, R. Amigó, A. García-Santiago, J. M. Hernández and J. Tejada, "Collective microwave emision from molecular clusters at low temperatures", *International Conference on Molecule-Based Magnets (ICMM 2004)*, Tsukuba, Japan, 4-8 October 2004. (Poster)
- M. Jordi, J. Tejada, R. Amigó, J. Vanacken, S. Stroobants, M. Malfait, V. V. Moshchalkov, E.M. Chudnovsky and D.A. Garanin, "Magnetization reversal in molecular nanomagnets under pulsed magnetic fields", *International Conference on Molecule-Based Magnets (ICMM 2004)*, Tsukuba, Japan, 4-8 October 2004. (Poster)

Prof. Javier Tejada gave the following invited talks:

- "Molecular Magnets and microwaves", City University of New York, 17 March 2004.
- "Molecular magnets and superradiance", New York University, 18 March 2004.
- "Microwave radiation from molecular clusters", *March Meeting 2004*, American Physical Society, Montreal (Canada).
- "The route from single domain particles to molecular clusters", University of Stuttgart, 16 July 2004.

#### CSIC:

N. Ventosa, "Producción de micro- y nano-partículas orgánicas utilizando CO<sub>2</sub> comprimido", I<sup>a</sup> Reunión Nacional de la Red Flucomp, Madrid (Spain) 12-14 November 2003.

- N. Ventosa, S. Sala, J. Veciana, "DELOS® Process: Crystallization of Pure Polymorphic Phases from CO<sub>2</sub>-Expanded Solutions", *6th International Symposium on Supercritical Fluids*, Versailles (France), 28-30 April 2003.
- M. Cavallini, M. Massi, F. Biscarini, J. Gómez, D. Ruiz-Molina, C. Rovira, J. Veciana, "Nanopatterning of molecular magnets by stamp-assisted self-organization", *E-MRS Spring Meeting*, Strasbourg (France), 9-13 June 2003.
- M. Cavallini, J. Gómez, M. Massi, F. Biscarini, D. Ruiz-Molina, C. Rovira, J. Veciana, "Deposition and nanopatterning of Mn<sub>12</sub> single-molecule magnets on surfaces with size and positional control at multiple length scales", *Trends in Nanotechnology*, Salamanca (Spain), 15-19 September 2003.
- J. Veciana, "Functional Supramolecular Nano-Architectures", Max Planck Institut, Stuttgart (Germany), 1 December 2003.
- Daniel Ruiz-Molina, "Isolated single-molecule magnets on the surface of a polymeric thin-film", Kinki University, Department of Chemistry, Osaka (Japan), 9 September 2003.
- Daniel Ruiz-Molina, "Isolated single-molecule magnets on the surface of a polymeric thin-film", Osaka University, Department of Chemistry, Osaka (Japan), 12 September 2003.
- D. Ruiz-Molina, J. Gómez-Segura and J. Veciana, "Multiple Length Scale Patterning of Single-Molecule Magnets", *1st NanoSpain Workshop*, San Sebastian (Spain), 10-12 March 2004. (Oral)
- D. Ruiz-Molina, J. Gómez-Segura and J. Veciana, "Multiple Length Scale Patterning of Single-Molecule Magnets", *III Reunión Nacional de Física del Estado Sólido*, San Sebastian (Spain), 2-4 June 2004. (Oral)
- <u>J</u>. Gómez-Segura, D. Ruiz-Molina and J. Veciana, "Nanopatterning of Single-Molecule Magnets on a surface", *IV Congreso Español de Fuerzas y Túnel*, Vic (Spain), 21-24 September 2004. (Oral)
- J. Gómez-Segura, D. Ruiz-Molina, C. Rovira and J. Veciana, "Multiple Length Scale Patterning of Single-Molecule Magnets", *Quantum Effects on Molecular Nanomagnets (QuEMolNa Kick-off meeting)*, Valencia (Spain), 28-29 May 2004. (Oral)
- J. Gómez-Segura, D. Ruiz-Molina, C. Rovira and J. Veciana, "Nanopatterning of Single-Molecule Magnets on a surface", 7th International Conference on Nanostructured Materials, Wiesbaden (Germany), 20-24 June 2004. (Poster)

- J. Gómez-Segura, D. Ruiz-Molina, C. Rovira and J. Veciana, "Nanopatterning of Single-Molecule Magnets on a surface", *International Conference on Molecular Magnets*, Tsukuba (Japan), 04-08 September 2004. (Oral)
- J. Veciana, "Functional Molecular Nano-Objects", *Jornades Franco-Catalanes de Nanociències i Nanotecnologia*, Barcelona (Spain) 5-6 July 2004.
- J. Veciana, "Nanopatterning of Mn<sub>12</sub> single-molecule magnets on surfaces. Towards magnetic information storage", Workshop on "Perspectives on Single-Molecule and Single-Chain Magnets" CREST, JST; Tsukuba (Japan), October 4th, 2004

#### NPL:

- L. Hao, J. C. Macfarlane, C. Gardiner, P. Josephs-Franks and J. C. Gallop, "Inductive Superconducting Transition-edge for Photon and macromolecule detection", *Condensed Matter* and Materials Physics Conference (CMMP03), 6-9 April 2003, Queen's University of Belfast, UK.
- L. Hao, "SQUIDs: Towards the nano-scale", 23 July 2003, invited talk at CSIRO, Lindfield, Sydney, Australia.
- L. Hao, J. C. Macfarlane, P. Josephs-Franks and J. C. Gallop, "Nanoscale Inductive Superconducting Transition-edge Photon and Particle Detector", *Nanoelectronics*, Lancaster University, Lancaster, UK, January, 2003 (poster).
- L Hao, J C Gallop, C. H. Gardiner, P. Josephs-Franks, J C Macfarlane, S K H Lam and C Foley, "Inductive superconducting transition-edge detector for single-photon and macro-molecule detection", 9th International Superconducting Electronics Conference 2003 (ISEC2003), 7-11 July 2003, Sydney, Australia (talk).
- P. Josephs-Franks, L Hao, A Tzalenchuk, J Davies, O Kazakova, J C Gallop, L Brown and J C Macfarlane, "Measurement of the spatial sensitivity of miniature SQUIDs using magnetic-tipped STM", 9th International Superconducting Electronics Conference 2003 (ISEC2003), 7-11 July 2003, Sydney, Australia (talk).
- A Tzalenchuk, "Feasibility of ultrasmall Josephson junctions for all-HTS phase qubits", *IOP Superconductivity Annual Conference*, Cambridge, UK, January 2003 (invited talk).
- A Tzalenchuk, "Dynamics and measurement issues of ultrasmall HTS Josephson junctions with

- unconventional current-phase relation", *Symposium on Quantum Measurements and Metrology*, Gothenburg, Sweden, September 2003 (invited talk).
- A Tzalenchuk, "Fabrication, dynamics, and possible qubit applications of ultrasmall HTS Josephson junctions with unconventional current-phase relation", *International Superconductivity Conference ISS'03*, Tsukuba, Japan, October 2003 (invited talk).
- 12th International Conference on Scanning Tunneling Microscopy/Spectroscopy and related techniques (STM03), 21-25 July 2003, University of Technology, Eindhoven (Netherlands)
- International Conference on Magnetism (ICM 2003),27 July 01 August 2003, Roma, Italy.
- Theoretical Trends in Low Dimensional Magnetism (LDM 2003), 23-25 July 2003, Firenze, Italy.
- L. Hao, J. C. MacFarlane, J. C. Gallop, and S. Lam, "Inductive sensor based on nano-scale SQUIDs", *Applied Superconductivity Conference 2004 ASC'04*, Jacksonville, Florida, USA, October 3-8 2004 (to be published).
- Condensed Matter and Materials Physics Conference (CMMP04), 4-7 April 2004, University of Warwick, UK.
- Conference on Precision Electromagnetic Measurements (CPEM 2004), 27 June 2 July 2004, London (UK).
- 4th International Symposium on Metallic Multilayers (MML04), 7-11 June 2004, Boulder, Colorado (USA).

#### HPLB:

- *1st QuICT Network meeting*, Abingdon, UK, July 2003. (Talk Quantum measurement in solid state quantum computers).
- 4th QIPC Workshop, Oxford, UK, July 2003 (1 poster Quantum measurement in solid state quantum computers).
- The International Conference on Solid State Quantum Information Processing (SSQIP), Amsterdam, The Netherlands, 15-18 December 2003 (1 talk, 3 posters).
- Few Qubits Applications of Quantum Information Processing, Budmerice, Slovakia, 11-14 December 2003 (two invited talks).
- T. M. Stace and S. D. Barrett, "Charge qubit Rabi oscillations in point contact detectors",

Quantum Dots Conference (QD2004), 10-13 May 2004, Banff, Alberta (Canada). (Poster)

- S. D. Barrett and T. M. Stace, "Continuous measurement of a microwave-driven solid state qubit", X International Conference on Quantum Optics (ICQO2004), 30 May – 3 June 2004, Minsk (Belarus). (Invited talk)
- S. D. Barrett and T. M. Stace, "Continuous measurement of a microwave-driven solid state qubit: an all-electrical analogue of resonance-fluorescence *Seventh International Conference on Quantum Communication, Mesaurement and Computing (QCMC2004)*, 25-29 July 2004, Glasgow (UK). (Talk)
- S. D. Barrett and T. M. Stace, "Techniques for spin measurement in exchange interaction quantum computers", *Seventh International Conference on Quantum Communication, Mesaurement and Computing* (QCMC2004), 25-29 July 2004, Glasgow (UK). (Poster)
- T. M. Stace and S. D. Barrett, "The role of inelastic tunnelling in a continuous quantum measurement of a solid state qubit", Seventh International Conference on Quantum Communication, Mesaurement and Computing (QCMC2004), 25-29 July 2004, Glasgow (UK). (Poster)
- B. W. Lovett, A. Nazir, S. D. Barrett, T. P. Spiller, and G. A. D. Briggs, "Selective Spin Coupling using a Single Exciton", *Mesoscopic Quantum Coherence and Computing*, 7-10 June 2004, Napoli (Italy). (Talk)
- S. D. Barrett and P. Kok, "Efficient high-fidelity quantum computation using matter qubits and linear optics", *QISW04* Cambridge (UK) (2004). (Poster)
- B. W. Lovett, A. Nazir, S. D. Barrett, T. P. Spiller, and G. A. D. Briggs, "Selective Spin Coupling using a Single Exciton", QISW04 Cambridge (UK) (2004). (Poster)
- W. J. Munro, K. Nemoto, T. P. Spiller, S. D. Barrett, R. G. Beausoleil, and P. Kok, "Linear and nonlinear quantum optical information processing". (Invited talk)

In addition to these international events listed above, HPLB members presented results at a number of smaller UK meetings (*UK EPSRC Quantum Circuits Network Meetings*, *Oxford Quantum Logic Gate Club Meetings*), and gave a number of seminars at various UK Universities and other institutions internationally, including the University of Queensland, the Centre for Quantum Computer Technology (University of New South Wales), and Harvard University.

Question 2.3.		
Invited naners muhlished in scientific or fechnical		Title and journals/conference and partners involved
journal or conference.	1	<u>UB:</u>
		• J. Tejada, R. Amigó, J. M. Hernandez, A. García-Santiago, "Resonant experiments in magnetism: superradiance and magnetic spectroscopy", ICM 2003 – International Conference on Magnetism, 27 July – 1 August 2003, Rome (Italy), Journal of Magnetism and Magnetic Materials 272-276 (2004) 2131-2135.
	3. Impa	npact on Innovation and Micro-economy
		A – Patents
Question 3.1.		When and in which country(ies):
Patents filed and pending	0	Brief explanation of the field covered by the patent:
Question 3.2. Patents awarded	0	When and in which country(ies): Brief explanation of the field covered by the patent* (if different from above):
Question 3.3. Patents sold	0	When and in which country(ies):  Brief explanation of the field covered by the patent* (if different from above):
Questions about project's outcomes	Number	Comments or suggestions for further investigation
		B - Start-ups
Question 3.4. Creation of start-up	No	If YES, details: - date of creation: - company name
		- subject of activity: - location: - headcount:

Question 3.5.  Creation of new department of research (ie: organisational change)	No	- turnover: - profitable : yes / no / when expected - web address:  Name of department and institution/company:	
C – Technology transfer of project's results			
Question 3.6.  Collaboration/ partnership with a company?	No	Which partner: Which company: What kind of collaboration?	
4. Other effects			
A - Participation to Conferences/Symposium/Workshops or other dissemination events			
Question 4.1.		Names/ Dates/ Subject area / Country:	
Active participation <sup>1</sup> to Conferences in EU Member states, Candidate countries / NAS. (specify if one partner or "collaborative" between partners)	1	HPLB:  • 4th QIPC Workshop organized by Quiprocone (www.quiprocone.org), Oxford, UK, July 2003 (Organizer)	
Question 4.2.  Active participation to Conferences outside the above countries (specify if one partner or "collaborative" between partners)	0	Names/ Dates/ Subject area / Country:	
B – Training effect			
Question 4.3.			

<sup>&#</sup>x27;'Active Participation' in the means of organising a workshop / session / stand / exhibition directly related to the project (apart from events presented in section 2).

Number of PhD students hired for project's completion	1	In what field:  Obtaining and processing of materials derived from Mn <sub>12</sub> molecules (1).
Questions about project's outcomes	Number	Comments or suggestions for further investigation
		C - Public Visibility
Question 4.4.  Media appearances and general publications (articles, press releases, etc.)	0	References:  HPLB:  T. P. Spiller, "Quantum Information Technology", <i>Materials Today</i> , 30 (January 2003).
Question 4.5.		(Please attach relevant information) References:
Web-pages created or other web-site links related to the project	2	<ul> <li>www.nanomagiqc.com</li> <li>www.quiprocone.org</li> <li>See also the webpage of each partner in the Consortium, available through the main webpage of the project (www.nanomagiqc.com).</li> </ul>
		(Please attach relevant links)
Question 4.6.  Video produced or other dissemination material	0	References:  (Please attach relevant material)
Question 4.7.  Key pictures of results	0	References:  (Please attach relevant material .jpeg or .gif)

D - Spill-over effects		
Question 4.8.  Any spill-over to national programs	Yes	<ul> <li>If YES, which national programme(s):</li> <li>DTI Quantum Metrology Programme</li> <li>Programa Nacional de I+D orientada (Tecnologías de la Información y las Comunicaciones), Plan Nacional de Investigación Científica, Desarrollo e Innovación Tecnológica 2000-2003, Ministerio de Ciencia y Tecnología, Spain.</li> </ul>
Question 4.9.  Any spill-over to another part of EU IST Programme	No	If YES, which IST programme(s):
Question 4.10.  Are other team(s) involved in the same type of research as the one in your project?	No	If YES, which organisation(s):