Generation of picosecond acoustic pulses in cobalt by ultrashort electron pulses in gold

Oleksandr Kovalenko[1], Viktor Shalagatskyi[1], Thomas Pezeril[1], Vitalyi Gusev[1], Denys Makarov[2], Luyang Han[2], Oliver G. Schmidt[2] and Vasily V. Temnov[1]

[1]Institut des Molécules et Matériaux du Mans, UMR CNRS 6283, Université du Maine, 72085 Le Mans cedex, France.
[2]Institute for Integrative Nanosciences, IFW Dresden, 01069 Dresden, Germany
oleksandr.kovalenko.etu@univ-lemans.fr

Abstract

Generation of ultrashort hot electron pulses in metals by femtosecond laser pulses [1] as well as the ultrafast electronic heat transport and energy transfer to the lattice, resulting into the emission of coherent acoustic pulses [2-4] were discovered two decades ago. However, mainly due to the technological limitations in sample fabrication, many fundamental questions such as the efficiency of heat transport by ballistic electrons as compared to diffusive transport remained unclear. Nowadays, the electronic properties of different metallic compounds based on hybrid metal-ferromagnet multilayer structures attract particular interest because of their applications in ultrafast spintronics [5] and magneto-plasmonics [6]. Very recently, ultrafast plasmonic studies allowed for a detailed characterization of ultrashort acoustic pulses generated by femtosecond laser pulses in hybrid gold-cobalt bilayer structures [7].

In order to investigate the ultrafast pathways of energy transport and conversion in these structures under femtosecond laser irradiation we have performed time-resolved pump-probe measurements. A set of hybrid multilayer samples composed of (111) gold layers with different thicknesses on top of a 30 nm-thin hcp-cobalt film deposited on a (0001) sapphire substrate has been manufactured by magnetron sputtering. Femtosecond p-polarized optical pump pulses (duration 100 fs, λ=800 nm, pulse repetition rate 500 kHz) were focused at the gold-air interface at oblique incidence (Fig. 1a). A weak time-delayed s-polarized ultrashort probe pulse (duration 100 fs, λ=400 nm) was used to record the dynamics of pump–induced reflectivity changes at the gold-air interface (Fig. 1b). At zero pump-probe delay time a strong electronic peak caused by free carrier absorption of pump photons in gold was observed in all investigated samples. Hot electrons excited in gold within the optical skin depth (13 nm) of pump light propagate across the sample at a velocity of the order of the Fermi velocity in gold, i.e. 1.4×10^6 km/s [1]. After crossing the gold layer hot electrons release their excess energy in the cobalt layer, where the electron-phonon coupling is much stronger and the electron mean free path much shorter than in gold. The transient temperature increase in cobalt is estimated to be at least one magnitude larger than in gold. Afterwards the thermal expansion of the cobalt layer generates an ultrashort compressional acoustic pulse propagating in both directions, into the sapphire and into the gold layer. The acoustic propagation in a (111) gold layer occurs at a speed of 3.45 km/s suggesting that the strain pulse should arrive at gold-air interface after the delay time of d/c_s. Figure 1b shows the acoustic echo in a sample with d=70 nm delayed by d/c_s~20 ps.

More physical insight can be gained from the detailed analysis of the acoustic echoes. Ultrafast acousto-plasmonic [6,7] and optical reflectivity measurements [8] demonstrated that optical techniques can be particularly useful to reconstruct the acoustic pulses with spatial extension smaller than the skin depth of probe radiation. In this case the time-derivative of the optical signal gives the pulse shape. For example, the time-derivative of the acoustic echo in the inset of Fig. 1b shows the bell-shaped acoustic pulse with the duration of 2 picoseconds. Taking into account that the speed of sound in hcp-cobalt is roughly 6 km/s, it gives the heat penetration depth in cobalt of $\delta_{cobalt}=\tau_{ac}c_s^{(Co)}$=12 nm. This value is in excellent agreement with the theoretically estimated length of hot electrons diffusion in cobalt and with most recent experimental observations [7].

We have observed the acoustic echoes in all investigated samples with d ranging from 70 to 270 nm. The acoustic pulse shape remained unchanged but strain amplitude decreased significantly. The dependence of strain amplitude on gold thickness in Fig. 1c shows an exponential decay with a decay length of 120 nm. The most recent theoretical investigation shows that the shapes of experimentally observed ultrashort electron pulses in gold [1] can be explained within the frame work of wave-diffusion theory [9]. The ballistic front is followed by the diffusive tail, which carries most of the pulse energy. Therefore the exponential decay of the acoustic amplitude with gold thickness can be adequately described by the conventional theory for hot electron diffusion in metals leading to the decay length of about 100-150 nm [3,4]. From the physical point of view this is a characteristic length of hot electrons diffusion in gold during the time of electron-phonon relaxation.

To summarize, using time-resolved pump-probe measurements we have observed the generation of ultrashort acoustic pulses in cobalt heated by ultrashort electron pulses excited in gold. These measurements provide 120 nm and 12 nm electronic heat diffusion lengths in gold and in cobalt, respectively. Given the case that illuminating the gold surface with circularly polarized light leads to the emission of partially spin-polarized photoelectrons [10], it remains completely unclear how long spin polarization can be carried by ultrashort electron pulses, a question crucial for applications in ultrafast spintronics [5].

References

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Figures

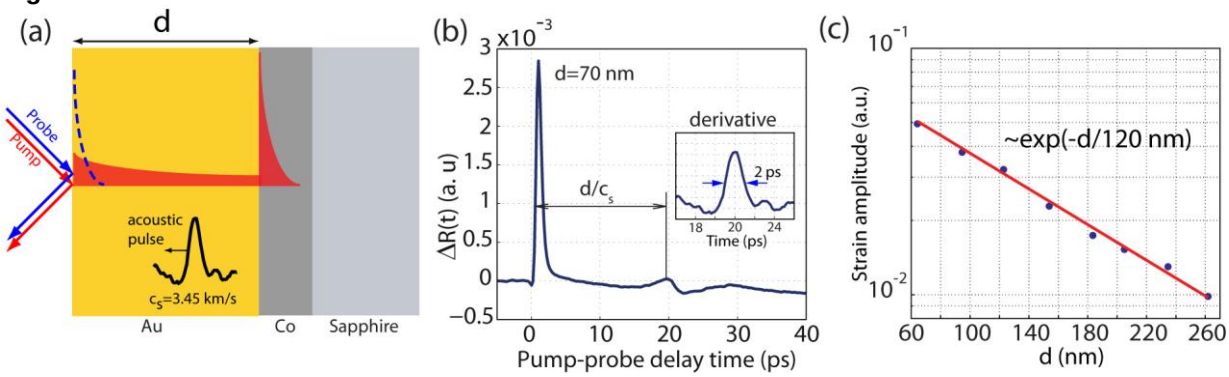

Fig. 1: (a) Schematic drawing of the femtosecond pump-probe experiment: hot electrons optically generated at the gold-air interface propagate through the layer of gold and heat up a 30 nm thin cobalt layer, resulting into generation of an ultrashort acoustic pulse. The red shaded area illustrates the distribution of lattice temperature in the gold/cobalt structure after electron-phonon relaxation (approximately 1 picosecond after pump excitation). The dashed curve shows the exponential decay of pump intensity within the skin depth. (b) Pump-probe reflectivity signal for a 70 nm thin gold layer. The inset shows the time-derivative of a time-delayed acoustic echo, which gives the acoustic pulse shape with a duration of 2 ps (see ref. [6,7] for details). (c) The exponential dependence of the acoustic strain amplitude on gold thickness.