Unusual spectral response of loss-compensated plasmonic nanoparticles in active gain media

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While based on phenomena recognized and described almost one and a half century ago [1], the physics of plasmons in metal nanoparticles has been recently fueled by the rapid development of new techniques for producing small particles and by the applicability of these structure in the realization of visible range metamaterials. One of the main issues in using metallic nano-structures for metamaterial applications at optical frequencies is their high level of losses. A most promising strategy to circumvent this obstacle is loss compensation, where the structures are coupled to active compounds (such as pumped dye molecules or quantum dots) which are able to transfer them energy and therefore amplify the desired response. Research along this line has recently gained momentum, resulting for example in the first demonstration of a nanoscale spaser using gain-assisted core-shell nanoparticles [2]. In this work, we study the apparently simple situation of a single metallic nanoparticle immersed in a gain medium with a focus on the plasmonic response, and show that interesting effects already arise with surprising modifications of the plasmonic spectral response.

We studied the behavior of a metallic nanosphere of radius *r* made of a metal of permittivity $\varepsilon_1 = \varepsilon_1' + i\varepsilon_1''$ (based on the experimental data from [3]) surrounded by an active (externally pumped) dielectric host with permittivity

$\varepsilon_2 = \varepsilon_2' + i\varepsilon_2''$ (with $\varepsilon_2'' < 0$ for gain). We here focus on the polarisability of the particle with respect to the outside medium. In the presence of gain, this is given in the quasi-static limit as:

$$\alpha=\alpha'+i\alpha''=4\pi r^3(\varepsilon_2'+i\varepsilon_2'')\frac{(\varepsilon_1'+i\varepsilon_1'')-(\varepsilon_2'+i\varepsilon_2'')}{(\varepsilon_1'+i\varepsilon_1'')+2(\varepsilon_2'+i\varepsilon_2'')}.$$

The plasmon resonance appears at the frequency ω_0 where $2\varepsilon_2' = -\varepsilon_1'$. As proposed in [4], perfect loss compensation is then obtained when the gain level is exactly adjusted ($2\varepsilon_2'' = -\varepsilon_1''$) at this same frequency: one then recovers a singular response. It is obviously highly desirable to obtain such a high amplitude plasmon in order to exacerbate the overall metamaterial response. However, our work points out that this singular behavior is intrinsically different from the ideal plasmon obtained from a lossless metal: in the latter case, the imaginary response $\alpha''(\omega)$ is a Dirac peak (resonant losses are confined to a very narrow spectral region), while in the perfectly-compensated plasmon $\alpha''(\omega)$ takes on a spectrally wide, $1/(\omega-\omega_0)$ behavior (Fig. 1-b and 1-f). To the best of our knowledge, this has remained unnoticed but has important implications, since it means that even with perfect loss compensation, resonant losses can be mitigated but they do *not* simply vanish away.

Moving away from the perfect compensation point by adding or removing gain, more unusual, and sometimes surprising features appear. Mathematically, this comes from the fact that the equations for the real and the imaginary part of polarizability are more similar in presence of external gain than in the absence of it, which gives rise to new behaviours. One striking example happens if, at the plasmon frequency, we have $\varepsilon_2''(\omega_0) = -\varepsilon_2'(\omega_0)$ as shown in Fig. 1-c and 1-e. In this situation, the real part $\alpha'(\omega)$ takes on a bell shape, while the imaginary part $\alpha''(\omega)$ has a zig-zag shape. This is exactly opposite to the usual plasmon case (Fig.1-a and 1-d) where – as is well-known from textbooks for any type of passive resonator – the *real* part should be zig-zagging and the *imaginary* part should be bell-shaped. We call this new behavior an “anti-plasmon”.

Beyond the peculiarity of this “anti-plasmon” behaviour, an interesting point is that where the real part of the polarisability is maximum, losses are zero (again in contrast with conventional plasmons, Fig. 1-a and 1-d). In Fig. 1-c, for example, one observes strongly negative response with low loss around the plasmon resonance, a property that could be most interesting if one is interested in metamaterials with negative properties based on such resonant elements. Note that the “anti-plasmons” can have either positive or negative real parts (Fig. 1-e and 1-c): this depends if, for the specific system considered, the condition for the anti-plasmon ($\varepsilon_2'' = -\varepsilon_2'$) is met at gain levels higher (Fig. 1, upper row) or lower (lower row) than the condition for perfect loss-compensation ($\varepsilon_2'' = -1/2\varepsilon_1''$).

Although our approach relies on a very simple theoretical description, it is worthwile noting that related behaviour appears, although left unnoticed by the authors, in much more sophisticated approaches (full FDTD simulations of split-ring resonators in active medium) [5].

Our work therefore underlines that the behaviours of loss-compensated plasmonic particles can be both unusual and richer than the usual case without gain, and that some of these could find, if confirmed in experiments, some potential applications in metamaterial designs.

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References

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Figures

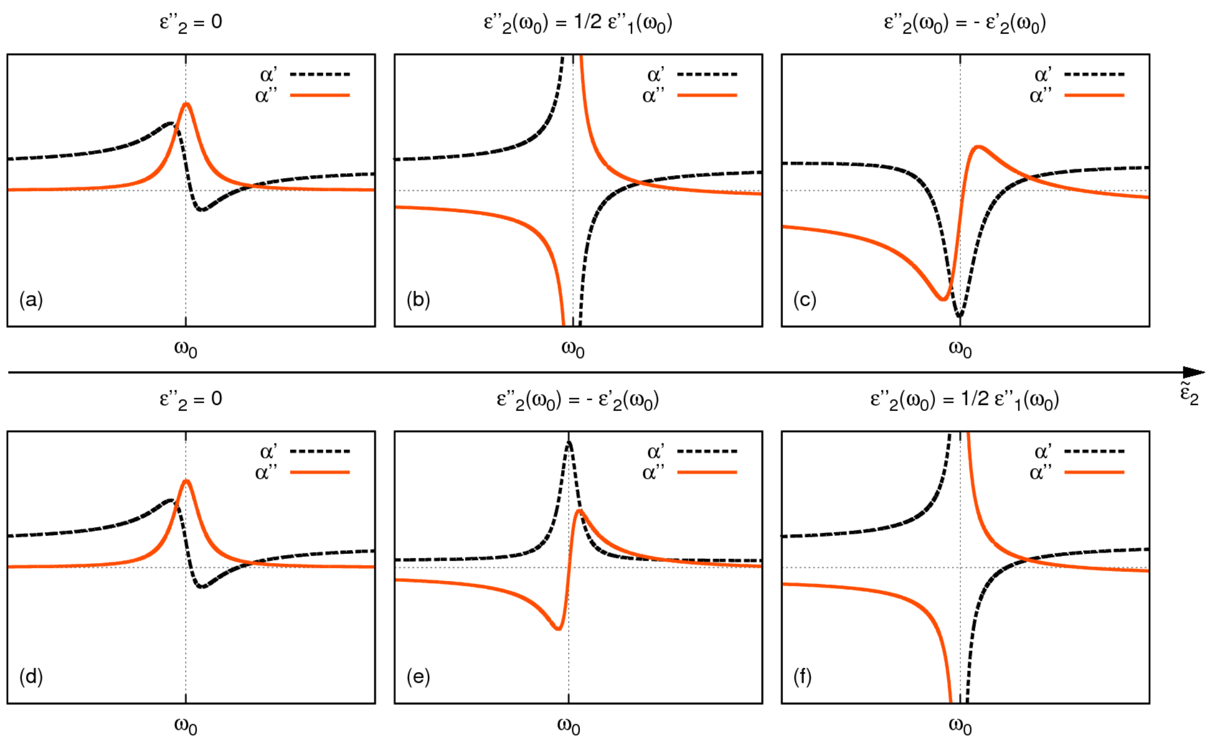

Fig. 1: Polarisability of a spherical nanoparticle in an active gain medium, with increasing gain levels from left to right. (a) and (d): Conventional plasmon resonance without gain. (b) and (f): when perfect loss compensation is obtained a $1/(\omega - \omega_0)$ behavior is obtained both for the real and the imaginary part. (c) and (e): when $\varepsilon_2''(\omega_0) = -\varepsilon_2'(\omega_0)$ an anti plasmon appear. The sign of the real part of the polarizability in a anti-plasmon resonance depends if it appears after the perfect loss compensation (a-c) or before it (d-f).