NONLINEAR OPTICAL RESPONSE OF DOUBLY-RESONANT ALUMINUM NANOCLOCKS

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The down-sizing of the active medium to the nanoscale makes their nonlinear optical responses less efficient. To overcome this low efficiency at the nanoscale, plasmon-enhanced nonlinear responses [1] using metallic nanoparticles have been proposed for nonlinear optics. The important aspect here is that the localized surface plasmon resonances (LSPRs) can concentrate incident light to strong local-fields ("hot spots") near the metal-dielectric interfaces. This is clearly favourable for driving nonlinear optical responses, which scale with a high power of the optical field.

In this context and as proposed previously [2], doubly-resonant structures are very promising for improving the nonlinear responses since they benefit from the field enhancement at both excitation and emission wavelengths. To meet this condition, the nanostructure or nanoparticle should be designed properly and the material well chosen. Here, we show numerically that double resonances can boost the second-harmonic generation (SHG) from a single aluminum (Al) nanoparticle. In order to avoid the limitations due to the oxidation, heating and dissipative effects in the most common plasmonic metals, e.g., gold and silver, recently Al has received wide interest as excellent material for plasmonics [3].

We have studied both the linear and SHG responses of nanoclocks. A particle consists of a disk with two different extended arms attached (e.g., nanorods). This structure allows getting double resonances: one at the fundamental wavelength and the second at the SHG wavelength. The numerical study highlights a particular geometry (75 nm for the first arm and 35 nm for the second arm and a disk with diameter of 100 nm) with a suitable linear response (around 1060 nm and 530 nm). We also verified that the local-field distributions of at the fundamental and SHG wavelengths are spatially mode-matched allowing efficient SHG radiation into the far field. Finally, the scattering cross section for SHG was computed. Our results show that the SHG scattering is significantly enhanced compared to a singly resonant particle when the fundamental and SHG wavelengths coincide with the two plasmon resonances of the NC.

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