Saturable and Reverse Saturable Scattering of a Single Gold Nanoparticle

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Nowadays, nonlinear optics plays an unprecedented role in research not only because of its intriguing physical mechanisms but the wide range of photonic functionalities, such as frequency generation, ultrafast signal processing, microscopy, etc. [1]. However, optical nonlinearities are usually weak. One of the most promising candidates is the nonlinearity of localized surface plasmon resonance (LSPR) in noble metal nanostructure. The strongly enhanced nonlinearity is usually attributed to the strong surface field enhancement due to the nanometer scale and the interband transitions near visible light. Many examples had been discovered, such as Kerr effect [2], saturable and reverse-saturable absorption (SA and RSA) [3].

Last year, we reported the saturable scattering (SS) of single gold nanosphere (GNS), showing great potential for bleaching-free super resolution microscopy [4][5]. Recently, we further discover the reverse-saturable scattering (RSS)[6]. In our experiment, a 561 nm CW laser is used as the light source of backward scanning confocal microscope, and the sample are isolated 80 nm GNSs immersed in oil, whose resonant peak is close to the laser. If we focus the laser beam at the center of GNSs with increasing intensities, the backward scattering will flatten at about 5 x 10^5 W/cm² (SS), decrease about one-third, start bouncing back sharply again at about 1.5×10^6 W/cm².

The phenomena are reminiscent of SA and RSA, where the nonlinear absorption of gold nanoparticles spread in dielectric material is measured with varying intensities using Z-scan method [7]. For SA and RSA, the nonlinearity is usually modeled by a three-level system, attributed to the finite number of electron in plasmon absorption, free-carrier absorption, and two-photon absorption [8][9]. In gold nanostructures, the scattering and absorption are closely linked via Mie theory. In addition, the intensity dependent curve of our scattering result is very similar to previous result of absorption. Therefore, the same mechanism that used to explain SA and RSA might also be used to explain SS and RSS.

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