

Study on the non-linear refraction of silver nanoparticles with aggregation effect

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Received 10 June 2004; received in revised form 24 September 2004; accepted 18 January 2005

Abstract

The non-linear refractive responses of a highly monodisperse 10.5 nm diameter silver nanoparticles with the addition of KNO₃ have been measured by Z-scan technique with 35-ps pulse-duration laser pulses at 1064 nm. Strongly enhanced non-linear refractions are observed by four times in the presence of KNO₃. Through the size distribution and transmission electron microscopy measurements, the morphology of the aggregated silver nanoparticles has been observed as a chain-like aggregation. The dramatically enhanced non-linear refractive response was explained by the enhanced electromagnetic field near the surface of the silver nanoparticles as they approached.

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PACS: 42.65.Hw

Keywords: Non-linear refraction; Silver nanoparticles; Z-scan and aggregation

1. Introduction

Noble metal nanometer-sized particles have attracted significant attention because of their unusual optical and electronic properties, and their potential applications in emerging optoelectronic and photonic technologies [1–6]. Their optical

properties including linear and non-linear responses have been extensively investigated [7–12]. The linear optical characteristics of them are recognized as being dominated by the surface plasmon resonance (SPR) that also enhances their non-linear optical response [5,8–10]. SPR is associated with the collective oscillation of particles' free electrons on conduction band accompanying enhanced local electromagnetic fields, which is intensively sensitive to boundary conditions. Therefore, the geometry of noble metal nanoparticles would provide important control over linear

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and non-linear optical properties [9]. Recently, some different methods have been developed to yield nanostructured materials with distinctive configuration and optical properties [13–15]. Aggregation could directly modify nanoparticles' geometry, and is convenient for comparing the experimental results with that of individual. The non-linear refraction of isolated silver nanoparticle and the influence of aggregate effect on second-order optical non-linearities have been reported [7,8,10]. Moreover, analysis of aggregates showed that the fundamental new characteristics were present in the two sphere dimer model [16]. Parallel to the second-order optical non-linearities [8,10], the non-linear refraction of aggregated silver nanoparticles has been relatively untapped, as we know. Accordingly, we expect the non-linear refractive response from aggregated silver nanoparticles to show an essential new feature.

In this paper, Z-scan technique, which has emerged over the past decade as a powerful method to determine the non-linear refraction of species [17,18], has been employed to study the non-linear refractive response from silver nanoparticle synthesized by wet chemical method as a function of KNO_3 . Here, non-linear refractive responses from aggregated silver nanoparticles were observed to be enhanced by 4 times. The characteristics of aggregated silver nanoparticles were studied by particle size measurements, transmission electron microscopy (TEM) and Uv-vis spectra. The dramatically enhanced non-linearity was explained by the enhanced electromagnetic field near the silver nanoparticle surfaces.

2. Experiments

An aqueous colloidal solution of spherical silver nanoparticles was synthesized following the literature procedure [19]. Briefly, 300 mg of AgNO_3 (ACROS) was added to 2000 ml of deionized water with stirring. After heating to 100 °C, a 40 ml aqueous solution of sodium citrate (China National Pharmaceutical Group Corporation) with 1% mass concentration was injected into the AgNO_3 solution. When the solution became straw yellow, heating was stopped, followed by cooling

the solution with water. The obtained yellow colloidal solution has a concentration of 8.8×10^{-4} M calculated in terms of the silver atom. Aggregation did not take place over 3-month time period.

The measurements of the experimental silver nanoparticles size were completed on a N4 PLUS type submicro particle size analyse which was produced by Beckman Coulter.

The non-linear refractive responses of the silver nanoparticles with various KNO_3 concentrations were studied by Z-scan technique. The experimental arrangement for Z-scan setup has already been described previously [17]. Briefly, a Gaussian distribution laser beam from a mode-locked Nd:YAG (Quantel YG901C), which provides 35-ps pulse-duration at repetition rate of 10 Hz and a wavelength of 1064 nm, was focused on the 1-mm-thick quartz cuvette by a lens with 30 cm focal length. The energy of the input laser pulse was about 10 μJ . The laser beam waist was about 51 μm at the focal point, and corresponding Rayleigh length was 1.9 mm that was larger than the thickness of sample. The aperture diameter was 1 mm and this corresponding to a far field transmittance of 1%. The input and output energies of the laser beam were measured by PIN photodiodes, which is connected with a signal integrator (Stanford Research Sys. SR250). Each data point was averaged over 30 shots. As a reference, we performed a Z-scan on CS_2 , and obtained $n_2 = 1.2 \times 10^{-11}$ esu, which is in a good agreement with the published result [17].

3. Results and discussion

Fig. 1(a) is the absorption spectrum for a freshly prepared silver colloidal solution. The maximum absorption at 420 nm known as the surface plasmon resonant peak represents the characteristic of the collective excitation of particles' free electrons on conduction band. About 80 nm full width at half maximum of plasmon resonant band suggests that the silver nanoparticles are well monodispersed [11,12]. This conclusion is consistent with the distribution result of the particle sizes, which is shown in Fig. 2. Figs. 1(b)–(d), are the absorp-

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