SECOND-ORDER NONLINEAR OPTICAL RESPONSE OF COLLOIDAL GOLD LABELED HUMAN SERUM IGG

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Abstract—Hyper-Rayleigh scattering (HRS) technique was used to study the second-order nonlinear optical properties of colloidal gold labeled human serum IgG. The HRS experiments performed on protein-colloid conjugates display strong HRS signal at 532 nm when pumped with high intensity laser pulse with a wavelength of 1064 nm. The measured signal has a quadric intensity dependence on the pump laser energy indicating that the signal measured is indeed due to a second-order nonlinear optical process, namely, hyper-Rayleigh scattering. Thus, apparently for the first time, the hyper-Rayleigh scattering from nanocrystals bioconjugates has been observed.

Key words: Hyper-Rayleigh scattering (HRS), gold colloids, human serum IgG, nonlinear optical property.

I. INTRODUCTION

Hyper-Rayleigh scattering (HRS) is a nonlinear incoherent light scattering process that has recently been employed to study the second-order nonlinear optical properties of molecules in solutions [1]. Briefly, HRS method relies upon the random fluctuations of the density or orientation of chromophores, which instantaneously break the centrosymmetry of the isotropic media and create conditions of net frequency doubling. Compared to the more traditional technique of electric-field-induced second harmonic generation, HRS method offers the advantage that it can be performed in a liquid phase without the need of application of an aligning electric field. Consequently, HRS has been successfully applied to study nonlinear optical properties of charged chromophores, non-dipolar chromophores and proteins dissolved in isotropic media [2-3]. Recently, enormous HRS signal from colloidal gold nanoparticles has also been reported [4].

In this paper we report the first experimental study of the second-order nonlinear optical properties of colloidal gold-labeled human serum IgG by using HRS technique.

II. EXPERIMENT

The HRS experimental setup used here is similar to that of Clays et al [1]. The light source was a Q-switched Nd:YAG laser (Continuum, Surelite II) with a pulse duration of 9 ns full width half maximum (FWHM), operating at a repetition rate of 5 Hz at a wavelength of 1064 nm. The pump laser beam was focused into a glass sample cell by a cylindrical lens system. The light was collected at 90° to the incident beam and was measured with a photomultiplier tube (Hamamatsu, R105UH). The signal was recorded with a digital oscilloscope (Tektronix TDS 3032). Wavelength discrimination was accomplished by means of a 3 nm bandwidth interference filter centered at 532 nm.

To prepare the aqueous gold colloids, 4 ml of 1% sodium citrate solution was added to 100 ml of 0.01% boiling tetrachloroauric acid solution. The mixture was stirred till deep wine red color was obtained indicating formation of colloidal gold suspension. TEM study reveals that the mean diameter of the gold particle is about 15 nm. The human serum IgG was prepared in our laboratory and the protein-colloid bioconjugates were prepared according to the modification of the literature [5]. Before HRS measurements, the colloidal gold labels IgG was diluted with distilled water to desired concentrations.

III. RESULT AND DISCUSSIONS

When pumped with high intensity laser pulse with a wavelength of 1064 nm, the colloidal gold labeled human serum IgG displays measurable signal at 532 nm. Typical temporal profiles of the measured HRS signals for colloidal gold labeled human serum IgG at various concentrations (number density) are shown in Figure 1. The width of the transient HRS signal is about 10 ns, which are mainly determined by the width of the pump laser pulse. The amplitude of the transient HRS signal are measured for various gold concentrations and plotted in the inset of Figure 1. It is clear that the HRS signal has a linear dependence on the sample concentration, which is in consistent with the theoretical prediction [1].

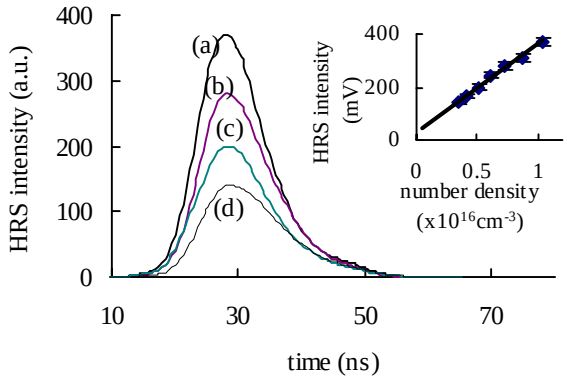

Figure 1 Temporal HRS signal profile of colloidal gold labeled human serum IgG at a number density of $3.5x10^{15}$ (a), $7.2x10^{15}cm^{-3}$ (b), $5.0x10^{15}cm^{-3}$ (b), $1.0x10^{16}cm^{-3}$ (d). The inset shows the linear dependence of the HRS signal intensity on sample concentration, over the range from $3.4x10^{15}$ to $1.3x10^{16}cm^{-3}$.

Semiconductor nanocrystals have been reported to be highly fluorescent and have been used as biological label [6]. In contrast, neither the nanocrystalline gold colloids nor the IgG molecules show measurable fluorescence signal, as studied by a fluorescence spectrophotometer (Shimadzu RF5000). Moreover, the measured HRS signal has a quadric intensity dependence on the pump laser energy. Therefore the signal measured here is indeed due to a second-order nonlinear optical process, namely, hyper-Rayleigh scattering. So far as we aware, this work represents the first experimental observation of hyper-Rayleigh scattering from protein-colloid bioconjugates.

Figure 2 shows the dynamic process of gold colloid and gold labeled IgG. The HRS signals from both bare gold colloids and protein-colloid conjugates first increase and then reach a stable value. However, the HRS signal from protein-colloid conjugates rises faster and has a larger stable value, as compared with that of bare gold colloids. We think that the enhancement is due to the change of the surface states of the gold nanocrystals. As we mentioned above, HRS is a second-order nonlinear optical process, which relies upon the none-centrosymmetry conditions of the molecules or nanoparticles. The modification of the gold nanocrystals with IgG molecules will change the surface states of the particles, and creates the none-centrosymmetry conditions. Therefore the HRS signals were enhanced. Hupp et al [4] also reported that the HRS signal is highly sensitive to the colloidal aggregates and surface conditions. We are currently developing a HRS biosensor based on this phenomenon.

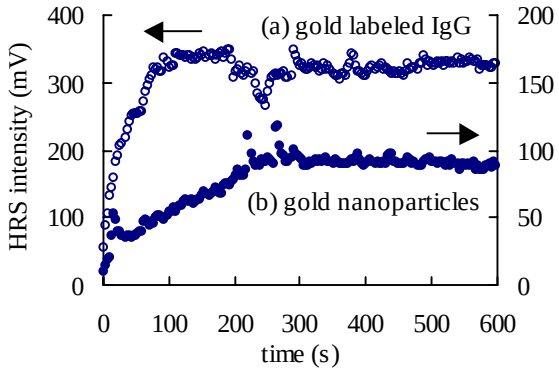

Figure 2 Dynamic process of colloidal gold and colloidal gold labeled human serum IgG.

IV. CONCLUSIONS

In summary, we reported here the first experimental observation of hyper-Rayleigh scattering from colloidal gold labeled human serum IgG. We believe that the experimental findings reported here have important biological and biomedical implications and should have high potential for further scientific and technological applications.

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