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Journal of Luminescence 119–120 (2006) 370–377
JOURNAL OF LUMINESCENCE
www.elsevier.com/locate/jlumin

Ultrafast nonlinear optical response of silver/bismuth oxide nanocomposite films with different silver concentrations

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Available online 23 February 2006

Abstract

The ultrafast nonlinear optical properties of co-sputtered silver/bismuth oxide ($Ag:Bi_2O_3$) nanocomposite films with different Ag concentration (13.2–59.3 at%) were investigated by femtosecond (fs) pump–probe and fs optical Kerr effect (OKE) techniques. The pump–probe measurements revealed the Ag concentration-related energy relaxation dynamics: for films with high concentration of Ag (>50%), the relaxation dynamics consist of a fast excitation power-dependent component (1–2 ps) and a long-lived component (>100 ps). While for films with relative low Ag concentration (<40%), the energy relaxation behaves as a single exponential process, nearly completed within 1 ps. This Ag concentration related dynamical behavior was suggested to be originated from the differences in the structural and thermal properties of both Ag nanoparticles and Bi_2O_3 matrices. The result of the femtosecond OKE measurements showed that the third-order susceptibilities ($\chi^{(3)}$) of $Ag:Bi_2O_3$ films have a maximum of 4.1×10^{-10} esu at Ag concentration of 35.7%.
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PACS: 78.67.Bf; 78.47.+p; 42.65.Pc

Keywords: Nanocomposite; Ultrafast; Nonlinear optical response; Optical Kerr effect

1. Introduction

The novel electronic and optical properties of noble metal nanoparticles, mainly determined by their size, shape and concentration, have drawn a great deal of attention in past decade [1]. The absorption spectra of metal nanoparticles in thin films or in colloidal solutions contain a characteristic absorption band in the visible region, so-called surface plasmon resonance (SPR) band [1,2]. The SPR arises from the surface plasmon oscillation modes of conduction band electrons in metal particles, and strongly influences the linear and nonlinear optical response of these nanoparticles.

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doi:10.1016/j.jlumin.2006.01.019

In the past years, several groups have studied the ultrafast electron dynamics of metal nanoparticles [3–7]. Studies revealed that the ultrafast excitations result in a non-Fermi distribution electron gas in metal particles, which thermalizes through electron–electron collisions (<500 fs) and electron–phonon coupling (0.5–5 ps) [4]. Changes of electron distribution during internal and external thermalization of the electron gas give rise to a modification of the dielectric function and optical response of the metal particle [3,4]. Recent studies have focused on the effects of surrounding medium on the overall energy relaxation process in composites containing metal nanoparticles, and found that the surrounding medium can play an import role in some cases [5,6].

Composite systems of noble metal nanoparticles embedded in a dielectric matrix often exhibit large third-order optical nonlinearities due to the enhancement of the local field near and inside the metal particles around SPR [8]. For instance, Liao et al. [9] reported very large $\chi^{(3)}$ in Au:TiO_2 composite films with high concentration (15–60%) of Au prepared by cosputtering, and a maximum value of $\chi^{(3)}$ was obtained at Au concentration of 38%. Solis et al. [10] also observed the similar dependence of $\chi^{(3)}$ on metal concentration in Cu:Al_2O_3 composite films. However, there were few reports about the nonlinear optical property of composite films with high concentration of Ag nanoparticle embedded in a dielectric matrix with high refractive index.

In this paper, we report our studies on the ultrafast nonlinear optical response of Ag:Bi_2O_3 nanocomposite films measured by femtosecond (fs) time-resolved pump–probe and fs optical Kerr effect (OKE) techniques. We found that the concentration of Ag has important influences on both the value of $\chi^{(3)}$ and the response time of optical nonlinearity of the composite films, which is determined by the excitation relaxation dynamics.

2. Experimental

2.1. Sample preparation

Ag and Bi_2O_3 were cosputterd by using multi-target magnetron sputtering method as described in detail in a previous paper [11]. The Ag and Bi_2O_3 targets were independently manipulated by DC and RF power supply, respectively. The relative composition of Ag in the composite film was determined by adjusting the power supply of Ag target, while the power supply of Bi_2O_3 target was fixed. The concentrations of Ag in these composite films were measured by using Rutherford back scattering (RBS) method. Composite films with Ag concentration of 13.2–59.3% were prepared by setting the power supply of Ag target at 20–150 W. The thickness of these composite films is about 120 nm determined by RBS measurements.

2.2. Fs pump–probe and OKE setup

The femtosecond dynamics measurements were performed in a standard pump–probe configuration. Pulses of 80 fs duration and 82 MHz repetition rate at 800 nm generated from a mode-locked Ti-Sapphire oscillator (Spectra Physics, Tsunami), were amplified by a regenerated amplifier (Spectra Physics, Spitfire) pumped by a Q-switched Nd:YLF laser. The amplified fs laser beam with 120 fs duration, 600 μJ pulse energy and 1 kHz repetition rate was split into two beams: the intense one with pulse energy about 400 μJ was used as pump source of an optical parametric amplifier (Spectra Physics, OPA-800CF) to produce tunable infrared laser pulses at wavelength from 1.1 to 1.5 μm, and the residual (200 μJ) was used as pump beam in two-color pump–probe measurements. The output of OPA was frequency doubled by using a BBO crystal to yield pulses at 550–750 nm, which were used in degenerate pump–probe measurement or used as probe beam in two-color measurement.

The femtosecond OKE experiments were performed using the output of oscillator stage. The set-up arrangement was similar to that of the pump–probe except that the polarization of the probe beam was set at 45° with respect to that of the pump beam, and an analyzer with orthogonal polarization to the probe beam was used to detect the OKE signal.

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