Magneto-optical Kerr effect in perpendicularly magnetized Co/Pt films on two-dimensional colloidal crystals

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Magneto-optical Kerr effect and optical reflectance are measured in the visible region for perpendicularly magnetized Co/Pt films on self-assembly array of two-dimensional closely packed polystyrene spheres. Peaks and valleys are observed in the magneto-optical Kerr and the optical reflectance spectra and their positions scale with the sphere diameter. In explanation of the above scenario, not only the Mie optical scattering of single nanocaps but also the surface plasmon resonance should be considered. This work will facilitate development of magnetoplasmonic nanosensors. © 2009 American Institute of Physics. [DOI: 10.1063/1.3182689]

Extraordinary optical transmission of light in an optically opaque metallic film perforated with a two-dimensional (2D) array of subwavelength holes was attributed to the coupling of surface plasmon resonance (SPR) to the light by Ebbeisen et al. Very recently, the effect of the SPR on magneto-optical Kerr effect (MOKE) has been studied extensively. It has been theoretically predicted that the MOKE can be enhanced greatly by the SPR. Alternatively, the large MOKE is desirable in practical applications. However, the MOKE of these plasmonic systems is usually smaller than that of corresponding continuous magnetic films. Therefore, it is required to further explore the MOKE in the plasmonic systems. Moreover, although perpendicular magnetic anisotropy is the prerequisite for applications of the magnetoplasmonic systems, most of the observed magnetic nanostructures almost have in-plane magnetic anisotropy.

In this letter, Co/Pt multilayers were deposited on self-assembly array of 2D closely packed polystyrene spheres (PSSs), where the Co/Pt films have perpendicular magnetic anisotropy and reasonably large MOKE.

The nanostructured Co/Pt films were prepared as follows: the silica slides were pretreated to render their surface hydrophilic by soaking in a solution of 30% hydrogen peroxide at 80 °C for 30 min and a U-shaped spacer was sandwiched by parallel silica slides to form a channel, which was in turn filled by an aqueous solution of colloidal dispersion with a suitable concentration. Here, the monodisperse PSSs (size dispersion 1%) were purchased from Duke Scientific Corps. After drying in air, highly ordered PSSs were grown within the channel, where the sphere diameter \( d = 400, 500, \) and 600. Afterward, Pt (15 nm) (bottom)/[Co(1 nm)/Pt(2 nm)]_1/Pt(10 nm) multilayers were then deposited by dc magnetron sputtering on the PSS arrays. Samples were characterized by x-ray diffraction (XRD), scanning electron microscopy (SEM), and atomic force microscopy (AFM). The polar Kerr rotation \( \theta_K \) and Kerr ellipticity \( \varepsilon_K \) were recorded by a magneto-optical Kerr spectrometer. Optical reflectance spectra were measured by microspectrophotometer and scanning spectrometer, where the reflectance was normalized to that of an Al mirror. All measurements were performed at room temperature.

The SEM image in Fig. 1(a) shows that the PSSs are densely packed in a hexagonal structure. In the upper part of the image, there is a vacancy through which the monolayer colloidal crystal substrate is demonstrated. For the Co/Pt nanocaps, the film thickness changes laterally on each sphere with the thinnest layer at the equator of each sphere, due to the locally curved surface of the spheres. In order to illustrate that the Co/Pt adjacent nanocaps are interconnected with each other, the samples were annealed at 150 °C for 1 h. With the melting point of about 130 °C, the PSSs are ex-
pected to dissipate after the heat treatment. Since the morphology almost does not change, as shown in Fig. 1(b), the metallic nanocaps are still connected at the equators of neighboring spheres. XRD measurements have shown that the Co and Pt layers in both the continuous film and the 2D Co/Pt nanocaps are polycrystalline with (111) preferred orientation. The typical polar Kerr loop in Fig. 1(c) shows that the 2D ordered Co/Pt nanocaps have perpendicular magnetic anisotropy due to the positive interface anisotropy between the Co and Pt layers with (111) preferred orientation.15

The $\theta_K$ and $\varepsilon_K$ spectra of the 2D ordered Co/Pt nanocaps are shown in Fig. 2. Following distinguished features are found. First, for the continuous films, $\theta_K$ and $\varepsilon_K$ change monotonically with wavelength. In contrast, for the 2D ordered Co/Pt nanocaps, $\theta_K$ and $\varepsilon_K$ both undergo several maxima and minima with wavelength. Second, at long wavelengths, the Kerr rotation of the 2D ordered Co/Pt nanocaps with small $d$ is enhanced, in comparison with that of Co/Pt continuous films. The nonmonotonic MOKE spectra are direct consequences of the 2D periodic structure. Finally, either $\theta_K$ or $\varepsilon_K$ minimum is redshifted with increasing $d$. In order to interpret above interesting results, the optical reflectance spectra of the samples were measured at normal incidence. Figure 3(a) shows the results of the 2D ordered Co/Pt nanocaps with different $d$. The reflectance has a minimum at 513, 624, and 747 nm for $d=400$, 500, and 600 nm, respectively. Figure 3(b) shows the calculated reflectance spectra by a finite-difference time-domain method,16 where the refractive index of the PSSs is assumed to be 1.59 in the visible region and that of the Co/Pt multilayers on silica substrate was measured by scanning ellipsometry.17 The calculated reflectance spectra reproduce the main features of the experimental results. In particular, the predicted minimum positions agree well with the experimental observations.

It is significant to compare the redshift in $\theta_K$, $\varepsilon_K$, and reflectance minima for the 2D ordered Co/Pt nanocaps. Figure 4 shows that the minima of $\theta_K$, $\varepsilon_K$, and reflectance are almost shifted in the same trend and that they are located at different wavelengths for the MOKE and the reflectance, possibly due to different wavelength dependence of the refractive index and of the magneto-optical constants of Co/Pt films. It is indicated that the structure observed in the MOKE spectra is correlated with that of the reflectance spectra as a function of $d$.15

It is necessary to elucidate the physical mechanism of the salient optical reflectance spectra. First of all, the spectral

FIG. 2. (Color online) $\theta_K$ (left column) and $\varepsilon_K$ (right column) spectra of the 2D ordered Co/Pt nanocaps with $d=400$ [(a) and (c)], 500 nm [(b) and (d)], and 600 nm [(c) and (g)]. For comparison, $\theta_K$ (d) and $\varepsilon_K$ (h) spectra of the Co/Pt continuous films are also given.

FIG. 3. (Color online) Measured [(a), (c), and (d)] and calculated (b) optical reflectance spectra of the 2D ordered Co/Pt nanocaps [(a), (b), and (d)], and of stochastic Co/Pt nanocaps and 2D ordered bare PSSs (c). In [(a) and (b)], $d=400$, 500, and 600 nm. In [(c) and (d)] $d=500$ nm. In [(a)–(c)], the incident angle is 0, and in (d) the incident angle varies as shown by the inset numbers. For clarification, the data are offset along the vertical axis. Here, the areal density of PSSs in the stochastic sample is about one-fifth of that of the 2D ordered one.

FIG. 4. (Color online) Wavelengths corresponding to $\theta_K$ (a), $\varepsilon_K$ (b), and reflectance (c) minima as a function of $d$. The inset numbers refer to the ordinal number of the reflectance minimum in Figs. 2 and 3(a).
structure is not induced by the intrinsic optical properties of the Co/Pt films because the reflectance of the continuous Co/Pt film changes monotonically with the wavelength. Second, the reflectance spectra of the 2D ordered Co/Pt nanocaps are not induced by the pure effect of the 2D periodicity. Figure 3(c) shows that the 2D ordered bare PSSs and Co/Pt nanocaps have different optical reflectance spectra with an identical \( d \).

As analyzed below, the effect of single Co/Pt nanocaps is one major reason for the optical reflectance of the 2D ordered Co/Pt nanocaps. When the size of the metallic spheres is smaller and larger than \( \lambda/10 \) (\( \lambda \) is the wavelength of light in vacuum), the extinction coefficient is mainly caused by the localized SPR and optical scattering, respectively. For the present Co/Pt nanocaps, the effect of the localized SPR can be neglected because ellipsometry measurements showed that for the Co/Pt multilayers \( \text{Re}(\epsilon_m) = -2 \) in the visible region and the condition of the localized SPR cannot be satisfied. Furthermore, with \( d \) close to \( \lambda \), the optical scattering plays a determinant role in the optical reflectance spectrum of single Co/Pt nanocap. Therefore, the reflectance spectra of stochastic Co/Pt nanocaps in Fig. 3(c) should mainly arise from the optical scattering. This can be further verified below. On one hand, the optical reflectance has a minimum near 600 nm and the parameter \( 2\pi d/\lambda = 5.23 \) with \( d = 500 \) nm and \( \lambda = 600 \) nm. On the other hand, according to the Mie scattering model, the first order extinction coefficient has a maximum under the condition of \( 2\pi d/\lambda = 5.20 \). Hence, the reflectance minimum for the stochastic Co/Pt nanocaps agrees well with the Mie scattering model. The relevance of the optical scattering is apparent in the low reflectance of the 2D ordered Co/Pt nanocaps near 600 nm. However, the optical reflectance spectra of the latter samples cannot be well explained merely by the optical scattering of single Co/Pt nanocaps.

For periodically perforated metallic films, the SPR can be excited by the incident light, thereby leading to the spectral structure. The precondition for the SPR excitation is satisfied by the interconnected Co/Pt nanocaps. Moreover, the Co/Pt film in the present work is reasonably thin for the SPR to occur at the metal/PSS interface because the SPR can be excited at the interface of metal/dielectric medium even for the metallic layer thicker than the corresponding optical penetration depth. The SPR is excited under the condition of \( k_z = k_0\sqrt{\epsilon_m}\gamma_{m,n} \), where \( k_{m,n} \) and \( \gamma_{m,n} \) are the wavevectors of the surface plasmon and the incident light along the film plane, and the reciprocal vector, respectively. One has \( k_{z} = \tilde{k}_0 \sqrt{\epsilon_m}\gamma_{m,n} \), where \( \tilde{k}_0 \) is the wavevector of light in vacuum, and \( \tilde{\epsilon}_z \) and \( \epsilon_m \) are the dielectric constants of the dielectric medium and the metallic film, respectively. Since the magnitude of \( \epsilon_m \) in the visible region is usually much larger than that of \( \epsilon_z \), \( k_{z} = \tilde{k}_0 \gamma_{m,n} \). For the closely packed hexagonal structure, \( G_{m,n} = 4\pi/\sqrt{3d}(mn+n\tilde{\gamma}) \) with integers \( m \) and \( n \). With the normal incident light, i.e., \( k_{0z} = 0 \), one has \( 2\pi d/\lambda = 4\pi/m^2+n^2/\sqrt{\epsilon_z} \). For a specific \( d \), the wavelength ratio of the first (\( m=1 \) and \( n=0 \)) to the second (\( m=1 \) and \( n=1 \)) order SPRs is theoretically expected to be \( \sqrt{2} \). For all three 2D ordered samples, the ratio is about 1.2. Considering the wavelength dependence of \( \epsilon_z \), the theoretical and measured values agree with each other, suggesting the importance of the SPR in the optical reflectance spectra in Fig. 3(a). This can be further proven by the redshift in the reflectance minimum with varying incident angle in Fig. 3(d), which is thought to be a fingerprint of the SPR. As shown below, the wavelengths for the reflectance minima cannot be described merely by the SPR and thus other factors should be considered. For the first order SPR, the theoretical value of \( 2\pi d/\lambda \) is 4.5 with the refraction index of the PSS of 1.59. From the results in Fig. 3(a), the observed value of \( 2\pi d/\lambda \) is about 4.90 for all three samples. This discrepancy may arise from the effects of the semishell shape of the Co/Pt nanocaps, the coupling of the SPRs at the bottom interface and the top surface, and the 2D ordered arrays of the triangle Co/Pt elements in the projected area of the pores between neighboring PSSs.

In conclusion, for the 2D ordered Co/Pt nanocaps with perpendicular magnetic anisotropy, the MOKE and the reflectance both undergo several minima in the visible region which are redshifted with increasing \( d \) almost in the same trend. The structures in the MOKE and optical reflectance spectra are accounted for by both the excitation of the SPR and the Mie optical scattering of single Co/Pt nanocaps. This work may facilitate developing magnetoplasmonic nanosensors with multiplexing capabilities.

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