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MAGNETISM AND FERROELECTRICITY

Mechanism of Magnetic Ordering in $Dy_{1-x}Ni_x$–Ni Bilayer Films

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Abstract—A mechanism for the magnetic ordering of dysprosium in $Dy_{1-x}Ni_x$–Ni bilayer films is proposed. This ordering was discovered earlier by the authors when studying magnetic circular dichroism. For x exceeding a threshold value (~0.05), the contribution from the $Dy_{1-x}Ni_x$ layer in a bilayer film to the magnetic circular dichroism over the temperature range 80–300 K is approximately equal in magnitude to the magnetic circular dichroism observed in a single-layer Dy film at temperatures below the ferromagnetic phase transition temperature of Dy (~100 K). Since magnetic circular dichroism is an effect linear in magnetization, the observed effect is associated with magnetic ordering of the $Dy_{1-x}Ni_x$ layer in bilayer films due to the simultaneous influence of two factors: the incorporation of Ni into the Dy layer and the influence of the continuous Ni sublayer. The ferromagnetic ordering of a dysprosium layer doped with nickel (under conditions of an atomic contact with a continuous nickel layer) was confirmed by the field dependences of the polar and longitudinal Kerr effects. It was shown that both layers in the bilayer structure are magnetized in the same direction and characterized by an anisotropy of the easy-plane type. The magnetic ordering is assumed to be due to the change in the density of states of the $Dy_{1-x}Ni_x$ alloy caused by hybridization with the narrow peaks near the Fermi level characteristic of nickel.

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1. INTRODUCTION

Metallic dysprosium is attracting research interest as a material that has an extremely large magnetic moment and a complicated magnetic phase diagram and is of potential use in nanostructures containing dysprosium in combination with other metals, semiconductors, or insulators. Despite the fact that the magnetic properties of Dy have been discussed in a number of classical works and monographs (see, e.g., [1–3]), new studies are performed leading to a better understanding of the magnetism of this metal [4]. In particular, in addition to the magnetic phases described earlier, namely, the paramagnetic (at $T > 175$ K), helical antiferromagnetic ($T_N > 175$ K), and ferromagnetic ($T_C = 85$ K) phases, a number of new phases (including the spin-flop and fan phases) were discovered in [4]. Due to the complicated phase diagram and the strong dependence of the phase transition temperatures on the crystal quality, the magnetic properties of dysprosium change significantly in going to small particles and thin film nanostructures. For example, the effect of the crystallite sizes and strains on the character of the phase transitions in fine-grained Dy samples was studied in [5]. In [6], it was shown that helical structures do not arise in Dy nanoparticles embedded in an aluminum film. In [7], it was demonstrated that the temperature T_C decreases or increases in Dy epitaxial films incorporated in Y–Dy–Y and Er–Dy–Er sandwiches and this effect was explained by the fact that the Dy single crystal is stretched (compressed) along the c axis because Y (Er) has a larger (smaller) lattice parameter.

The magnetic state of a Dy layer in film structures or superlattices can be influenced by the interaction with the neighboring 3d-metal layers or inclusions of 3d-metal atoms in the Dy layer. There are many publications devoted to the study of layered Dy–3d-metal structures (see, e.g., [8–10]). Earlier, we discovered a strong influence of low Ni-impurity concentrations (~0.05) in Dy layers on the temperature and spectral dependences of magnetic circular dichroism (MCD), linear in magnetization, in $Dy_{1-x}Ni_x$–Ni bilayer films [11]. Over the temperature range 80–300 K, a Dy layer containing a Ni-impurity concentration x less than several mass percent was found to make a temperature-independent contribution to the MCD, with this contribution being approximately equal in magnitude to the MCD of a $Dy_{1-x}Ni_x$ single layer observed only below the ferromagnetic phase transition temperature T_C. This behavior of the MCD with variations in temperature was explained by ferromagnetic ordering of dysprosium caused by two factors: (i) Ni atoms distributed over the entire Dy layer thickness and (ii) the spin system of the adjacent transition-metal layer.

In this work, we study the field dependences of the polar and longitudinal Kerr effects (PKE and LKE, respectively) and propose a mechanism for the influence of Ni on the magnetic state of a Dy layer.

2. EXPERIMENTAL TECHNIQUES AND RESULTS

The samples for study were prepared on a modernized Angara molecular-beam epitaxy device by sputtering Ni and Dy on glass substrates at 250°C. The sputtering rates were ~0.05 and 2 Å/s for Ni and Dy, respectively. First, a Ni layer was deposited on a substrate, and then Ni and Dy were sputtered simultaneously. Three samples were obtained in a single cycle: Ni and $Dy_{1-x}Ni_x$ single layers and a Ni–$Dy_{1-x}Ni_x$ bilayer film. The component distributions over the surface and thickness of a sample were determined using Auger electron spectroscopy.

The magnetic state of samples was studied predominantly with the MCD method, because, first, this method is very sensitive and, second, there is no contribution from substrates to the MCD. The MCD was measured over the spectral range 350–650 nm as the difference $\Delta D = D_+ - D_-$ between the optical densities of a sample for waves having right-handed and left-handed circular polarization, respectively, with respect to the external magnetic field. The magnetic field was directed normal to the sample plane, and its strength was 4.5 kOe. The MCD was measured with an accuracy of $\pm 10^{-4}$. The samples were cooled in a nitrogen gas flow cryostat over the temperature range 80–300 K, with the temperature being maintained accurate to within ±1 K. At room temperature, the field dependences of the LKE and PKE were measured in in-plane magnetic fields of up to 4.0 kOe and in magnetic fields of up to 14 kOe directed normal to the sample plane, respectively.

Figure 1 shows the Auger profiles of Ni–Dy and Ni–$Dy_{1-x}Ni_x$ bilayer films. In the former case, there are no Ni inclusions everywhere in the Dy film except in the ~30-Å-thick interface, whereas in the latter case Ni is distributed uniformly over the entire thickness of the Dy layer. The Ni concentration was varied from 0.005 to 0.06. The problem of Dy oxidation was considered in detail in [11], where it was shown that Dy is in the metallic state everywhere in the layer except in the ~40-Å-thick surface layer. This conclusion is confirmed by the resistivity value (~5×10^{-5} Ω cm) and a high optical absorption (~10^{-5} cm^{-1}).

In [11], we presented the spectral dependences of the MCD of Dy and Ni single layers at 93 and 300 K and showed that, at 93 K, the MCD of Dy was comparable in magnitude to that of Ni but was opposite in sign. As the measurement temperature was increased to 300 K, the MCD of the Dy single layer decreased by one order of magnitude. The specific features of the MCD spectra of Dy are in reasonable agreement with experimental measurements [12, 13] and calculations [14] of the optical conductivity of bulk Dy. Analogous results are obtained for $Dy_{1-x}Ni_x$ films and for the Dy layers in Dy–Ni bilayer films (after the MCD of the Ni layer is subtracted from the measured overall MCD).

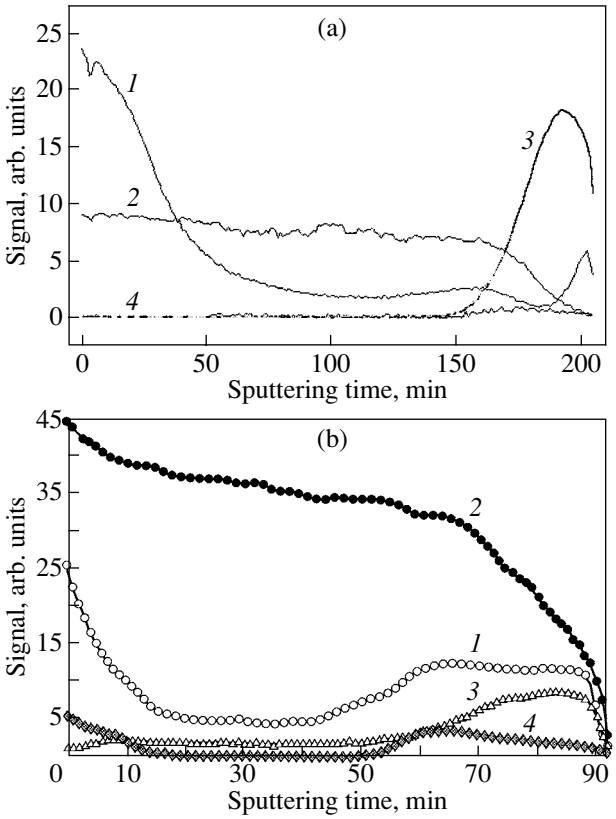

Fig. 1. Auger profiles of the main-element distributions over the thickness of bilayer films: (a) Ni–Dy; electron beam energy, 3 keV; current, 10–40 nA; ion sputtering at an energy of 3 keV and a current of 400 nA; and (b) Ni–$Dy_{0.95}Ni_{0.05}$; electron beam energy, 3 keV; current 100 nA; ion sputtering at an energy of 3 keV and a current of 600 nA. The curves correspond to the following elements: (*1*) oxygen, (*2*) dysprosium, (*3*) nickel, and (*4*) carbon.

Figure 2 shows the temperature dependences of the MCD of Ni, Dy, and $Dy_{1-x}Ni_x$ single layers and Ni–Dy and Ni–$Dy_{1-x}Ni_x$ bilayer films. The light wavelength is the same in all cases and is equal to 525 nm. For the Dy and $Dy_{1-x}Ni_x$ single layers, the curves are close to each other and are similar in character to the temperature dependence of the magnetization of fine-grained Dy samples studied in [5]. An order-of-magnitude increase in the MCD is observed near ~120 K, which corresponds to the transition of Dy into the ferromagnetic state. As in [5], there is no specific feature near T_N. The MCD of the Ni–Dy bilayer film is the sum of the effects exhibited by the Ni and Dy single layers. For the Ni–$Dy_{1-x}Ni_x$ bilayer films, the situation is completely different: for x exceeding the threshold value ~0.05, the temperature dependence of the MCD is similar to that for a Ni single layer but the magnitude of the MCD is close to the sum of the MCD magnitudes exhibited by Ni and Dy single layers at $T < T_C$. Thus, the temperature dependence of the MCD is unusual in this case. One might suppose that Dy has an effect on the MCD of Ni and decreases its magnitude. However, we carried out a

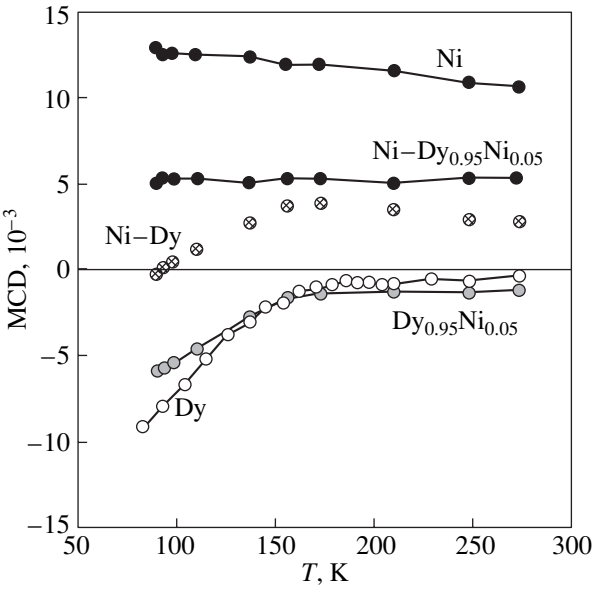

Fig. 2. Temperature dependences of the MCD in Ni, Dy, $Dy_{0.95}Ni_{0.05}$, Ni–Dy, and Ni–$Dy_{0.95}Ni_{0.05}$ films (magnetic field, 4.5 kOe; wavelength, 525 nm).

special experiment on Dy oxidation and found that the complete oxidation of the Dy layer (a reference Dy single layer was oxidized to Dy_2O_3) under the condition that the Ni layer remains in the metallic state causes the MCD magnitude to recover to that of a reference Ni single layer. The considerable contribution from the $Dy_{1-x}Ni_x$ layer to the overall MCD of the Ni–$Dy_{1-x}Ni_x$ bilayer film may be due to ferromagnetic ordering of this layer at temperatures far exceeding T_C of the corresponding single layer. The sign of this contribution is the same as that of the MCD of Dy at $T < T_C$. Based on this fact, we can assume that the magnetic moments of the Ni and $Dy_{1-x}Ni_x$ layers are parallel to each other.

In contrast to the MCD, which is the integrated effect exhibited by both layers of a bilayer film, measurements of the LKE and PKE characterize either of these layers separately. Figures 3a and 3b show the field dependences of the LKE for a Ni–$Dy_{0.95}Ni_{0.05}$ bilayer film as observed from the sides of the Ni and $Dy_{1-x}Ni_x$ layers, respectively. In both cases, there is a hysteresis loop, which is characteristic of the ferromagnetic state. The LKE for the $Dy_{1-x}Ni_x$ layer is opposite in sign to that for the Ni layer, as is the case with the MCD. For Dy and $Dy_{1-x}Ni_x$ single layers, the LKE is absent at room temperature. Figure 3c shows the field dependence of the PKE for the case where light is incident on the $Dy_{1-x}Ni_x$ layer of a bilayer film. In addition to the signal from the upper layer, there is a very weak signal of opposite sign from the lower Ni layer, which accounts for the shape of the observed curve. In the case of oblique incidence of light on a bilayer, the LKE signal from the lower layer is not observed for an *s*-polarized wave due to the strong reflection of the *s* component from the front surface. Hysteresis is absent. This fact, as well as the strong difference between the saturating magnetic fields for the LKE and PKE, indicates that both layers have easy-plane anisotropy and that the layers differ significantly in terms of their magnetization.

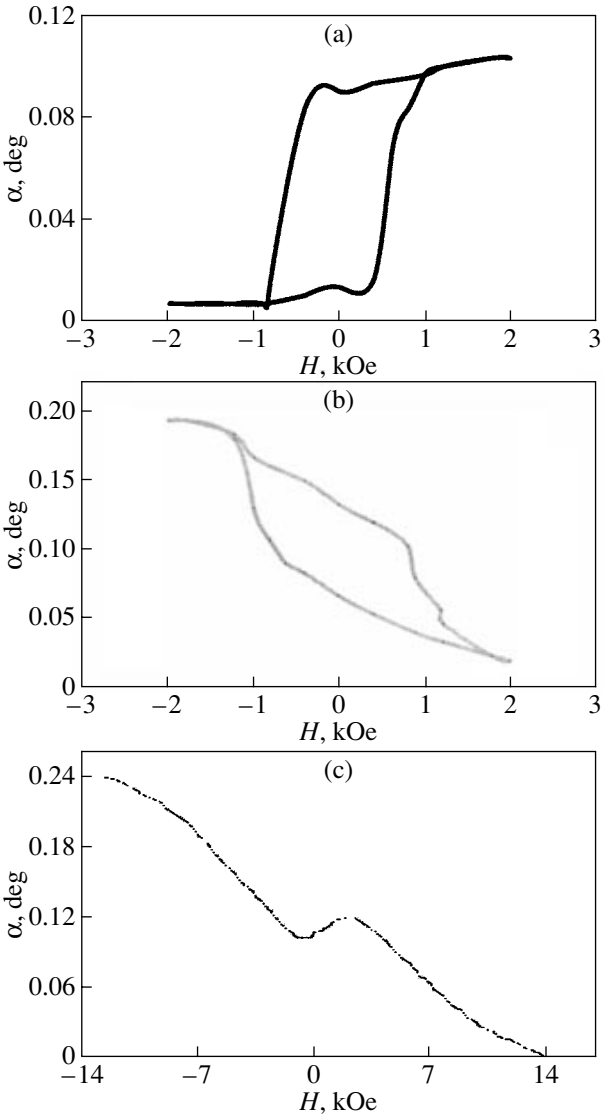

Fig. 3. Field dependences of the Kerr effect in a Ni–$Dy_{0.95}Ni_{0.05}$ film: (a) LKE for light reflected from the Ni layer, (b) LKE for *s*-polarized light wave reflected from the $Dy_{0.95}Ni_{0.05}$ layer (wavelength, 450 nm; angle of incidence, 45°), and (c) PKE for light reflected from the $Dy_{0.95}Ni_{0.05}$ layer (wavelength, 500 nm; normal incidence).

3. DISCUSSION OF THE RESULTS

The observed field dependences of the PKE and LKE are inconsistent with the assumption that the magnetic ordering of the $Dy_{1-x}Ni_x$ layer is magnetostatic in nature. Moreover, if the Dy layer had been simply magnetized by the magnetic field of the Ni layer, then this

would also have occurred in a Ni–Dy bilayer film, which was not the case. It also does not seem likely that the magnetic state of the $Dy_{1-x}Ni_x$ layer in bilayer films changes due to magnetostriction via the mechanism proposed in [7], because these films are not epitaxial. If the change in the structural parameters causing the magnetic ordering had been due to incorporated Ni atoms, an analogous effect would also have been observed in $Dy_{1-x}Ni_x$ single layers, which was not the case.

The magnetic polarization of the 4*f* electrons of rare-earth metals on a 3*d*-metal surface has been studied theoretically and observed experimentally in very thin layers. For example, in [15], the polarization of 4*f*-electron spins was observed in monatomic Dy layers deposited on a single crystal or a 3*d*-metal film. The spins of Dy atoms were antiparallel to the spins of the surface atoms or the 3*d*-metal layer. The fact that the Ni concentration has a strong effect on the temperature dependence of the MCD of a $Dy_{1-x}Ni_x$ layer suggests that, in this case, inclusions of Ni atoms and the connection of these inclusions to a magnetically ordered continuous Ni layer are responsible for the expansion of the Ni influence on the magnetic state of Dy deep into the Dy layer.

It is well known that the magnetic structure of heavy rare-earth metals (including dysprosium) is mainly determined by the exchange interaction of the conduction electrons with 4*f* electrons [4]. The incorporation of Ni atoms into a Dy layer causes changes in the system of conduction electrons. We propose the following model for these changes. In the spirit of the virtual-crystal model, we can write the density of states of the $Dy_{1-x}Ni_x$ alloy in the form

$$N(E) = (1-x)N_{\mathrm{Dy}}(E) + xN_{\mathrm{Ni}}(E).$$

The density of states of Dy near the Fermi level is dominated by the 5*d* electrons with a small admixture of 6*s* and 6*p* electrons [16, 17]. In Ni, there is a narrow filled peak of spin-up electronic states and a narrow unfilled peak of spin-down electronic states [18]. The density of states of the alloy is shown schematically in Fig. 4, where the Fermi levels of both alloy components are assumed to be equal. In the presence of a Ni layer, Ni atoms diffuse deep into the alloy and the Ni 3*d* electrons cause the Fermi level to rise. As a result, the Fermi level coincides with the spin-down electron peak and the $N(E_F)$ value increases. When a critical concentration is reached, the Stoner criterion can be satisfied and the Ni–$Dy_{0.95}Ni_{0.05}$ system can acquire magnetic properties at room temperature. In the absence of an additional Ni layer, the density of states at the Fermi level does not increase. In a Ni–Dy bilayer film, Ni 3*d* electrons also pass into the Dy layer; however, the specific feature in the density of states is absent in pure Dy and, therefore, the magnetic properties of a Ni–Dy bilayer differ only insignificantly from those of Dy.

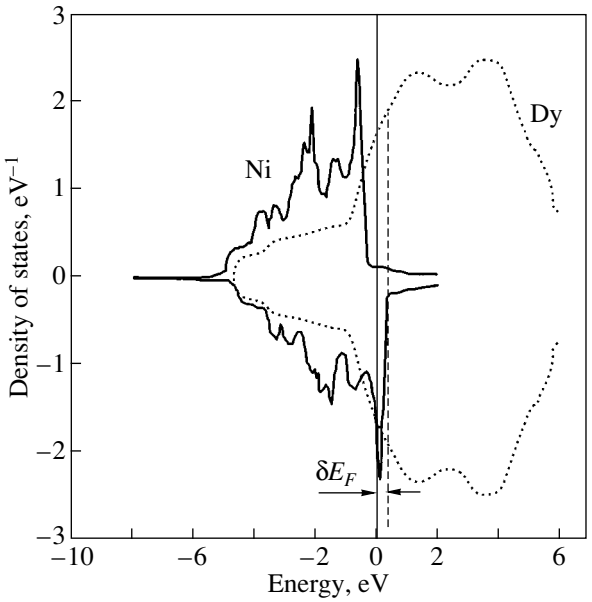

Fig. 4. Schematic diagram of the density of states of Dy (dotted curve) and Ni [18] (solid curve). The shift in the Fermi level caused by the penetration of electrons from the Ni layer is indicated.

4. CONCLUSIONS

Thus, as a component of the alloy, Ni causes a peak in the density of states to appear near the Fermi level. When Ni atoms diffuse from the lower layer, this peak is filled and the magnetic properties of the upper layer change if the Stoner criterion is satisfied.

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