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Characterization, transport, and magnetic properties of Co/Pd multilayers produced by pulsed laser deposition

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Metallic Co/Pd multilayers with equal layer thickness and periods ranging from 5 to 150 Å were deposited using pulsed laser deposition. They have an fcc (111) texture and the diffraction patterns give clear evidence of a modulated period structure. From the period dependence of the in-plane electrical resistivity a diffuse interface between cobalt and palladium of thickness \( b = 6.4 \pm 2 \) Å has been deduced. Magnetic, magnetoresistance, and Kerr-effect measurements indicate in-plane magnetization. In low fields \((B_0 < 10 \text{ mT})\) the magnetoresistance is highly anisotropic, varying by \(-2\%\) in the transverse configuration and \(+1\%\) in the longitudinal configuration due to orbital moment of cobalt in the interface region. The shape of the Kerr-effect major hysteresis loop can be explained by the in-plane component of the anisotropy of the cobalt layers.

I. INTRODUCTION

At present metallic magnetic multilayers are a subject of active research. Recent discoveries such as the giant magnetoresistance, the oscillation of the magnetic interaction between coupled ferromagnetic layers, and the large magneto-optical effects have stimulated many studies in the last few years. Many questions still arise concerning magnetism and the contribution of the interface to the resistivity. The origin of the multilayer phenomena is still not clear for many systems.

In this paper we study the transport and magnetic properties of Co/Pd multilayers produced by pulsed laser deposition, which is a new method for preparing metallic multilayers.

The dependence of the in-plane electrical resistivity on the multilayer period has been investigated and the magnetoresistance studied. Many authors discuss resistivity data within the framework of the Fuchs-Sondheimer size effect theory. This theory predicts an unphysical divergence of the resistivity for small periodicities, which is weakened, but not removed, by introducing an empirical "specular reflection factor." Here we consider the dependence of the electrical resistivity on the thickness and the profile of the diffuse interface. A simple interdiffusion model is used to explain the observed resistivity dependence and deduce the thickness of the interface layer. The strongly anisotropic magnetoresistance is analyzed in terms of anisotropic spin-orbit scattering and is related to the magnetization curves measured by Kerr effect.

II. EXPERIMENTAL PROCEDURE

The pulsed laser deposition technique uses a high-power laser to vaporize source material. The deposition of the multilayers was carried out in a HV chamber at a base pressure of \( 2 \times 10^{-7} \text{ mbar} \). A KrF excimer laser (Lambda Physik EMG 102 MSC) with a 248-nm wavelength, a 23-ns pulse width, and an average fluence of 10 J/cm\(^2\) was used, operating at a repetition rate of 10 Hz. Equal-thickness Co and Pd alternating layers were deposited at room temperature on fused silica amorphous substrates.

The deposition rate of about 0.5 Å/s was measured in situ using an oscillating quartz monitor that was used to evaluate period and thickness of the films. Multilayers with periods ranging from 5 to 150 Å were prepared and the total film thickness varied between 0.05 and 0.2 µm.

Scanning electron microscopy (SEM) was used to observe the surface morphology of the films. The average composition was obtained by energy-dispersive x-ray analysis (EDX). The structural characterization of the layered films was done by x-ray diffraction on a Siemens D500 diffractometer using Cu Kα radiation.

The in-plane electrical resistance of the multilayer samples was measured by the standard four-point method. Current flow was in the plane of the multilayers. The temperature dependence was determined between 295 and 10 K in a closed-cycled refrigerator. Magnetoresistance was measured in the three geometries: longitudinal (magnetic field and current parallel), transverse (field in the sample plane perpendicular to the current), and perpendicular (field perpendicular to the sample plane). The magnetoresistance is defined as \( \Delta R (B) / R(0) \), with \( \Delta R (H) = R (B) - R (0) \), and \( R (B) \) the resistance at an applied field \( B \). Measurements were carried out at room temperature in a field of up to 1.4 T and at 4.2 K in a field of up to 6 T. The hysteresis loops of the multilayers with an applied field of up to 0.3 T were obtained by magnetooptical Kerr effect operating in the transverse geometry.

III. RESULTS AND DISCUSSION

A. Structure

All laser-ablated Co/Pd multilayers are smooth and have a metallic luster. The SEM photographs show the presence of some metal droplets on the surface. An average chemical composition of approximately 50 at. % Co and 50 at. % Pd was determined from the EDX analysis. The structural analysis shows an fcc (111) Pd texture that is compatible with either (111) fcc or (002) hcp for Co. The fcc and hcp structures differ only in the layer stacking, ABCABC for fcc and ABAB for hcp. Thus to determine the structure, at least three Co layers are necessary. Here
FIG. 1. $\theta$-$2\theta$ high-angle x-ray diffraction patterns for equal-layer-thickness Co/Pd multilayers: (a) $\Lambda=153.1$ Å, (b) $\Lambda=51.6$ Å, (c) $\Lambda=47.2$ Å, (d) $\Lambda=29.5$ Å, (e) $\Lambda=19.0$ Å, and (f) $\Lambda=5.5$ Å.

we maintain the notation fcc (111) as a convention. Samples of large period $\Lambda$ show only the (111) reflections of Pd and Co. With decreasing $\Lambda$ the diffraction patterns are characterized by the evolution of satellites around these (111) reflections (Fig. 1). Low-angle diffraction measurements also confirm a periodic layered structure. The x-ray diffraction peaks are reasonably sharp and the linewidth does not show any obvious dependence on period. This indicates good periodic superlattices but tells us nothing about interface alloying since for statistically periodic disorder all peaks remain sharp.

B. Resistivity

The dependence of the electrical resistivity on $\Lambda$ (Fig. 2) indicates deviations from the thin-film size effect theory for small periods. Model calculations that assume interdiffusion at the Co/Pd interface predict that the resistivity approaches a plateau value for small periods. The formula

$$\rho(\Lambda) = \rho_{\text{Co}} + \rho_{\text{Pd}} + (4b\rho_{\text{M}}/\Lambda)\tanh(\Lambda/4b),$$

(1)

where $\rho_{\text{Co}}$ and $\rho_{\text{Pd}}$ are the bulk resistivities of Co and Pd, respectively, $\rho_{\text{M}}$ is the plateau resistivity, and $b$ is the thickness of the diffuse interface, can be used to deduce $b$. The experimental data clearly indicate a resistivity plateau at small $\Lambda$ (Fig. 2). Fitting of the data yields a thickness of the interface $b=6\pm2$ Å. Impurity scattering at the interface, and hence $\rho_{\text{M}}$, depends on the spin orientation of the scattering atoms, which is a likely explanation for the isotropic giant magnetoresistance in diffuse multilayers and granular systems.

C. Magnetoresistance

Magnetoresistance measurements and Kerr-effect hysteresis loops indicate in-plane magnetization in the as-prepared state for all the Co/Pd multilayers investigated here. The behavior of the magnetoresistance at low field is quite different in the three experimental geometries used (Fig. 3). A maximum negative value $\Delta R(II)/R(0)$ of about $-2\%$ at 4.2 K has been measured for the $\Lambda=29.5$ Å multilayer in the transverse geometry, whereas the longitudinal and perpendicular geometries magnetoresistance is positive, +1% and +0.5%, respectively. The low-field hysteresis clearly demonstrates the micromagnetic origin of the magnetoresistance. When the magnetic domain structure is eliminated the resistivity approaches a constant value.

Anisotropic magnetoresistance is a spin-orbit effect and can be written as

$$\rho(\Lambda) = \rho_{\text{Co}} + \rho_{\text{Pd}} + (4b\rho_{\text{M}}/\Lambda)\tanh(\Lambda/4b).$$

(2)

FIG. 2. Normalized (reduced) resistivities for Co/Pd and Pd/Au multilayers. Dashed line: Fit according to the size-effect theory. Solid line: Fit corresponding to Eq. (1) with $b=4.9$ Å. Pd/Au data are taken from Ref. 7.

FIG. 3. Magnetoresistance of a $\Lambda=29.5$ Å Co/Pd multilayer at 4.2 K. Measurements in the three geometries: (a) transverse, (b) perpendicular, and (c) longitudinal.
\begin{equation}
\rho(\theta) = \rho_1 + \Delta \rho_0 \cos^2 \theta,
\end{equation}

where $\theta$ denotes the angle between the current and the magnetization. For random three-dimensional orientation of the spins the resistivity is given by $\rho=\rho_1 + \frac{1}{2}\Delta \rho_0$, whereas random two-dimensional orientation yields $\rho=\rho_1 + 0.7\Delta \rho_0$, which may be explained by a sixfold in-plane anisotropy. The hysteresis in the magnetoresistance curves in Fig. 3 indicates local free-energy minima at $\theta=0$ and $\theta=\pi/2$. Both minima are separated by a free-energy barrier, and therefore it is incorrect to describe the anisotropy simply as easy-plane.

It is known that cobalt impurities in palladium contribute a positive anisotropic magnetoresistance of 1\%-7\%, which is associated with a large orbital moment of $0.7 \mu_B$ on the cobalt impurity. In our multilayers, there will be two or three layers of cobalt with many palladium neighbors in the diffuse interface region. It is the orbital moment on these atoms that is most likely the source of the observed magnetoresistance anisotropy.

Figure 4 shows the Kerr-effect hysteresis loop of the $\Lambda=20.2$ Å Co/Pd multilayer. The in-plane magnetic field is applied in an arbitrary direction. Subsequent turning of the magnetic field by an angle of $\pi/2$ yields minor loops with small hysteresis, which is interpreted in a way analogous to the perpendicular magnetization in uniaxial ferromagnetic crystals. The magnetic free energy in the plane is

\begin{equation}
F = -K'_3 \cos[6(\phi-\phi_0)] - HM_0 \cos \phi,
\end{equation}

where $\phi_0$ is assumed to be random and $K'_3$ is the in-plane anisotropy constant of the fcc (111) plane, which has been used to reproduce the major hysteresis loop. The value of $K'_3$ is 0.46 K J/m$^3$. To calculate Fig. 4, only irreversible contributions (switching fields) have been taken into account. The quality of this approximation, which is reasonable for major loops only, can be estimated from the remanence $M_R = M_0$, which may be compared to the exact value $M_R = 0.95M_0$.

IV. CONCLUSIONS

Good-quality Co/Pd metallic multilayers can be prepared by pulsed laser deposition. The period dependence of the resistivity yields the thickness of the diffuse interface between Co and Pd of about 6 Å. Anisotropic magnetoresistance is attributed to anisotropic spin-orbit scattering related to the orbital moment of the cobalt atoms in the interface region. The shape of the in-plane hysteresis loop can be explained by a sixfold in-plane anisotropy, and the constant $K'_3 = 0.46$ K J/m$^3$ is determined.

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