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Magnetic linear dichroism of infrared light in ferromagnetic alloy films

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We have observed a magnetic linear dichroism effect for infrared light in ferromagnetic alloys at room temperature. The effect has been studied for $Ni_{80}Fe_{20}$, $Ni_{80}Co_{20}$, $Co_{90}Fe_{10}$, and $Fe_{88}V_{12}$ thin films by measuring the transmission of light polarized parallel and perpendicular to the magnetization direction. Its physical origin and frequency dependence are explained from a two-current Drude-type model for the conductivity. A study of a single (thin film) specimen is shown to be sufficient for obtaining the spin and angular dependence of electron relaxation times in ferromagnetic alloys. [S0163-1829(99)50134-3]

The electrical conductivity of ferromagnetic materials is often viewed as resulting from the sum of parallel contributions of electrons with opposite spin directions. This so-called two-current model has provided excellent descriptions of the dependence of the residual resistivity of dilute binary and ternary ferromagnetic alloys on composition[1,2] and of the giant magnetoresistance of metallic multilayers with the current perpendicular to the planes (CPP-GMR).[3,4] Both types of studies yield the separate majority- and minority-spin resistivities of ferromagnetic metals, $\rho^{\uparrow}$ and $\rho^{\downarrow}$. However, knowledge of the spin-resolved resistivities is insufficient to describe transport in thin films with the current in the plane (CIP) of the film. In this so-called CIP geometry transport effects depend critically on the spin-dependent electron mean free paths, $\lambda^{\uparrow}$ and $\lambda^{\downarrow}$, in each of the layers that comprise the film. Model treatments of the CIP-GMR effect in multilayers, almost invariably using the free-electronlike Drude model, have been employed to extract $\lambda^{\uparrow}$ and $\lambda^{\downarrow}$ from the experimental data.[5–8] Room-temperature experiments give conflicting results as to whether or not there is spin-dependent bulk scattering in ferromagnetic materials.[7,8] It would be of much interest to be able to critically test the validity of this model for ferromagnetic materials, and to be able to explicitly obtain the spin-dependent mean free paths and relaxation times.

In this article we report on the discovery of a magnetic linear dichroism effect in ferromagnetic alloys at room temperature, and on its analysis on the basis of a Drude-type two-current model containing a small set of parameters. For thin alloy films we have observed a wavelength dependent difference between the transmission of infrared light that is polarized perpendicular or parallel with respect to the (in-plane) magnetization direction of the films, and provide strong evidence that the effect is related to the dc anisotropic magnetoresistance (AMR) of these films. The relationship is in a certain sense analogous to that between the recently discovered magnetorefractive effect in $(Ni_{80}Fe_{20}/Cu/Co/Cu)_N$ multilayers and the GMR effect,[9] however with the crucial difference that the latter effect is polarization independent. Analyses of the magnetorefractive effect in multilayers have been carried out using relaxation times averaged over all layers.[9] In contrast, studies of the magnetic dichroism of single layers avoid this averaging problem, making it possible to obtain material-specific relaxation times.

We remark that the linear dichroism effect reported here is of an entirely different origin than the Cotton-Mouton effect, observed for nonmagnetic materials in a transverse magnetic field.

We have studied four different ferromagnetic materials: $Ni_{80}Fe_{20}$ and $Co_{90}Fe_{10}$, which are frequently used in magnetoelectronic devices, $Ni_{80}Co_{20}$, which has a higher AMR ratio than $Ni_{80}Fe_{20}$, and $Fe_{88}V_{12}$, which was selected because of the reported qualitatively different angular dependence of the spin-dependent conductivity as compared to the other materials mentioned.[2] The films were fabricated using dc magnetron sputtering with a background pressure of 3×10^{-4} Pa, an Ar pressure of 0.9 Pa during deposition, and a substrate-target distance of 7 cm. On Si substrates polished on both sides a 3 nm thick Ta buffer layer was deposited, which induces a strong (111) texture in the fcc-type magnetic layers. Then the ferromagnetic layer was deposited, with a thickness that ranged between 3 and 50 nm. Finally, a protective Ta cap layer with a thickness of 3 nm was deposited. All layers were deposited at room temperature. The resistance and the AMR ratio of the films were measured using four-point probe methods. The film thicknesses were determined with Rutherford backscattering spectroscopy, assuming bulk densities.

The infrared transmission was measured using a Bio-Rad 175C spectrometer equipped with a mercury-cadmium-telluride (MCT) detector at normal incidence of the light. We measured in a wavelength range 2.5–20 μm (4000–500 cm^{-1}) with a resolution of 8 cm^{-1}. A polaroid filter with an efficiency of 98.5% was placed between the sample and the detector so that only light with a fixed linear polarization was detected. The magnetization of the films was saturated by the application of a magnetic field and could be rotated to any arbitrary angle θ with respect to the polarization angle which was kept fixed. The sample chamber was flushed with nitrogen gas to reduce the influence of water vapor and CO_2 on the transmission spectrum. The transmis-

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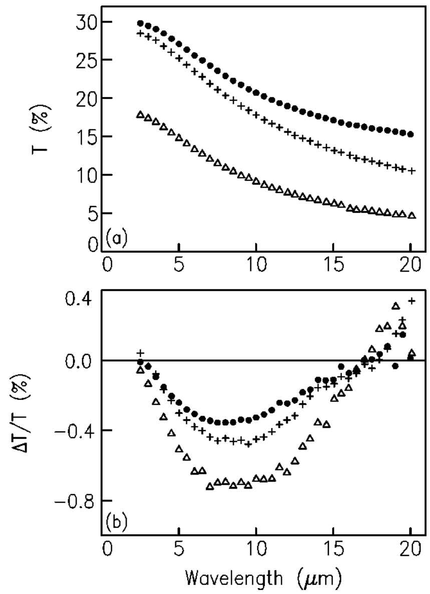

FIG. 1. Transmission (a) and magnetic linear dichroism effect (b) as a function of wavelength for 8 nm (●), 11 nm (+), and 19 nm (△) $Ni_{80}Fe_{20}$ layers.

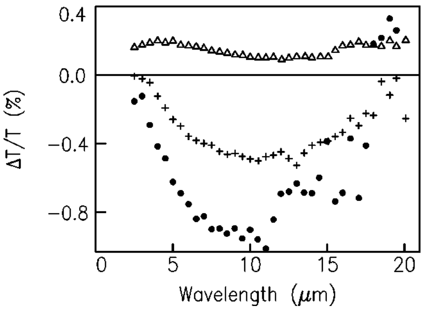

FIG. 2. Magnetic linear dichroism effect for 18 nm $Co_{90}Fe_{10}$ (+), 26 nm $Ni_{80}Co_{20}$ (●), and 10 nm $Fe_{88}V_{12}$ films (△).

sion coefficients given are normalized with respect to the transmission through an uncovered Si substrate. All experiments were carried out at room temperature.

Figure 1 gives an overview of results obtained for $Ni_{80}Fe_{20}$ films. As shown in Fig. 1(a), the transmission decreases monotonically with increasing wavelength and film thickness. The curves shown here have been measured with the magnetization direction perpendicular to the polarization direction. Figure 1(b) shows the linear dichroism effect, i.e., the relative change of the transmission, $(\Delta T/T)(\theta) \equiv [T(\theta) - T(90°)]/T(90°)$, with θ the angle between the magnetization and polarization direction, at $\theta = 0°$. The linear dichroism effect is small but significant. It increases with increasing film thickness until a maximum is reached at approximately 19 nm $Ni_{80}Fe_{20}$. The $\Delta T/T$ curve shows a minimum at a wavelength of approximately 8 μm. For wavelengths above 15 μm the statistical errors are relatively large due to the low amount of transmitted light in this wavelength range. The linear dichroism effect, $(\Delta T/T)(\theta)$, is found to decrease monotonically at each wavelength from $\theta = 0°$ to $\theta = 90°$ with increasing θ, similar to the dc AMR effect. Within experimental error $(\Delta T/T)(\theta = 0°) = 2 \times (\Delta T/T)(\theta = 45°)$.

As shown in Fig. 2 the effect is also observed for the other alloys studied. For $Co_{90}Fe_{10}$ and $Ni_{80}Co_{20}$ the wavelength dependence of the linear dichroism effect is similar to that of $Ni_{80}Fe_{20}$, but the size of the effect is smaller and larger, respectively. In contrast, the small effect observed for $Fe_{88}V_{12}$ films shows a markedly different wavelength dependence.

The observation of a magnetic linear dichroism effect implies that the complex refraction index depends on the angle θ between the polarization and magnetization direction. The complex refraction index is given by

$$\eta = \sqrt{\epsilon_r - \frac{i\sigma(\omega,\theta)}{\epsilon_o \omega}}, \tag{1}$$

with ϵ_o the dielectric constant in vacuum, and ϵ_r and $\sigma(\omega,\theta)$ the relative dielectric constant and conductivity of the metal. The effect on η due to the displacement of electrons in closed shells, as represented by $\epsilon_r \approx 1-10$, is very weak for the highly conducting films used in our study, and as the results of our analysis do not strongly depend on ϵ_r, we will neglect this term from now on.

We model the effect in terms of a frequency and angular dependent conductivity $\sigma(\omega,\theta)$, which is the sum of contributions from majority- and minority-spin electrons:

$$\sigma(\omega,\theta) = \sum_{\uparrow,\downarrow} \frac{n^{\uparrow(\downarrow)} e^2 \tau^{\uparrow(\downarrow)}(\theta)/m^{\uparrow(\downarrow)}}{1 + i\omega\tau^{\uparrow(\downarrow)}(\theta)}, \tag{2}$$

in which n and m are the spin-dependent effective electron density and mass, respectively. It is assumed that the conduction electrons behave as a free electron gas. The relaxation time τ is given by

$$\tau^{\uparrow(\downarrow)} = \tau_o^{\uparrow(\downarrow)}(1 - a^{\uparrow(\downarrow)} \cos^2 \theta). \tag{3}$$

We will use the parameters $a^{\uparrow}$, $a^{\downarrow}$, and the ratio $\alpha = \tau_o^{\uparrow}/\tau_o^{\downarrow}$ to fit the $\Delta T/T$ curves as will be shown later. It is assumed that $\sigma(\omega,\theta)$ is constant over the film thickness.[10] The form of Eq. (3) leads to a $\cos^2\theta$ dependence of the dc AMR effect, which, to a good approximation, is also the angular dependence of the linear dichroism effect as found from this model.

The frequency of the infrared light is so low that interband transitions can be neglected, and only intraband excitations are assumed possible. The transmission coefficients of the total layer stack are calculated by assuming continuity of the electromagnetic fields at the interfaces.[11] Including multiple internal reflections did not result in large changes of the calculated linear dichroism effect and will be neglected. For both Ta layers we have assumed a refractive index independent of angle and frequency in the experimental wavelength range, resulting in a frequency independent transmission. We have observed that the variation of the transmission through a single Ta film over this wavelength range is indeed small enough to justify this assumption.

From the measured dc resistivity at $\theta = 90°$, together with the measured curves of $\Delta T/T$, the parameters $\tau_o^{\uparrow}$, $\alpha \equiv \tau_o^{\uparrow}/\tau_o^{\downarrow}$, $a^{\uparrow}$, and $a^{\downarrow}$ are determined. We have assumed that the Ta cap layer is totally oxidized and therefore nonconducting. A reasonable estimate for the spin-independent relaxation time in the Ta seed layer is approximately 1

TABLE I. Total film resistivity, ρ, measured (calculated) AMR ratio, and value of $\tau_o^{\uparrow}$, α, $a^{\uparrow}$, and $a^{\downarrow}$ as obtained from a fit of the $\Delta T/T$ curve (see text) for $Ni_{80}Fe_{20}$, $Ni_{80}Co_{20}$, $Co_{90}Fe_{10}$, and $Fe_{88}V_{12}$ films of different thickness d.

	d (nm)	ρ (nΩ m)	AMR % exp.(calc)	$\tau_o^{\uparrow}$ (fs)	α	$a^{\uparrow}$	$a^{\downarrow}$
$Ni_{80}Fe_{20}$	8	290	1.3 (0.8)	6.7	1.5	0.035	−0.033
	11	230	1.7 (1.4)	8.5	1.5	0.056	−0.050
	19	210	2.1 (2.1)	11	2.5	0.040	−0.027
$Ni_{80}Co_{20}$	26	170	3.6 (3.6)	14	2.8	0.060	−0.031
$Co_{90}Fe_{10}$	18	160	1.3 (1.3)	16	3.5	0.021	−0.014
$Fe_{88}V_{12}$	10	200	0.5 (0.2)	15	11	0.001	0.007

$\times 10^{-15}$ s. This relaxation time is so low that also the influence of the Ta seed layer can easily be neglected. In a first analysis of the experimental data it was found that, when assuming $m^{\uparrow}=m^{\downarrow}=m_e=9.1\times 10^{-31}$ kg, and $n^{\uparrow}=n^{\downarrow}$, the best fits were found for an electron density $n^{\uparrow}=n^{\downarrow}=1.08\times 10^{28}$ m^{-3} and $n^{\uparrow}/m^{\uparrow}=n^{\downarrow}/m^{\downarrow}=1.19\times 10^{58}$ $m^{-3}\,kg^{-1}$ (resulting in $v_F=1.0\times 10^{6}$ m/s). The results of the fits are given in Table I, together with the AMR ratio that follows from these parameters, using $\Delta R/R=\alpha a^{\uparrow}+a^{\downarrow}/\alpha+1$ at $\omega=0$. This expression for $\Delta R/R$ results from Eqs. (2) and (3), including the spin-asymmetry parameter α. The low measured resistivities are indicative of the good structural quality of the films. The experimental AMR ratio and the value obtained from the fit parameters are in fair agreement, for $Ni_{80}Fe_{20}$ in particular, for the thickest films, for which the effect of the presence of the Ta layer is less pronounced. The linear dichroism effect as well as the AMR effect increase with thickness (up to a certain maximum). This is most likely the result of the decreasing importance of spin-independent scattering at the interfaces with the Ta layers, and/or at grain boundaries or other defects within the $Ni_{80}Fe_{20}$ film.[12] For $Ni_{80}Fe_{20}$, $Co_{90}Fe_{10}$, and $Ni_{80}Co_{20}$, it is found that $a^{\uparrow}$ and $a^{\downarrow}$ have opposite sign, whereas for $Fe_{88}V_{12}$ $a^{\uparrow}$ and $a^{\downarrow}$ have equal sign. These results are in qualitative agreement with the conclusions of Dorleijn,[2] who determined the values of the angular dependencies using resistivity measurements on dilute ternary Ni-based and Fe-based alloys. Note that the use of the present method is not restricted to dilute systems. The parameter $n^{\uparrow}/m^{\uparrow}$ (for $n^{\uparrow}/m^{\uparrow}=n^{\downarrow}/m^{\downarrow}$) can only be varied in a small range ($\pm 5\%$) around the values used above, when requiring a good fit to the experimental $\Delta T/T$ curve, the AMR ratio, and the experimental film resistivity. The resulting relative uncertainties of the parameters α, $a^{\uparrow}$, and $a^{\downarrow}$ are approximately 10%.

In Fig. 3 the experimental transmission and linear dichroism effect of a 19 nm $Ni_{80}Fe_{20}$ film are given together with the fits obtained from the model. It is shown that the wavelength dependence of $\Delta T/T$ can be explained quite well, and that the transmission above 7 μm wavelength is well described by the model, but that the fit is not satisfactory at shorter wavelengths. Figure 3 also shows the transmission change that is obtained when either $a^{\uparrow}=0$ or $a^{\downarrow}=0$, i.e., when either majority or minority electrons do not exhibit an angular dependence of the relaxation time. When $a^{\uparrow}$ and $a^{\downarrow}$ are small (<0.1), as is the case in the example shown, the total transmission change is to a good approximation the sum of the separate curves. This result shows immediately the effect of the two separate spin directions on the total linear dichroism effect.

Subsequent analysis of the data showed that when allowing $n^{\uparrow}/m^{\uparrow}\neq n^{\downarrow}/m^{\downarrow}$, a much larger range of values is possible for these parameters. However, in the analysis it is found that for every material investigated there is an approximate linear relationship between the values of $n^{\uparrow}/m^{\uparrow}$ and $n^{\downarrow}/m^{\downarrow}$, for which the best fit to all experimental results is obtained. For a 19 nm $Ni_{80}Fe_{20}$ layer the fit is good for $0.5\times 10^{58}<n^{\downarrow}/m^{\downarrow}<2.5\times 10^{58}$ $m^{-3}\,kg^{-1}$ when $n^{\uparrow}/m^{\uparrow}=-0.30\times n^{\downarrow}/m^{\downarrow}+1.54\times 10^{58}$ $m^{-3}\,kg^{-1}$. At the same time, α re-

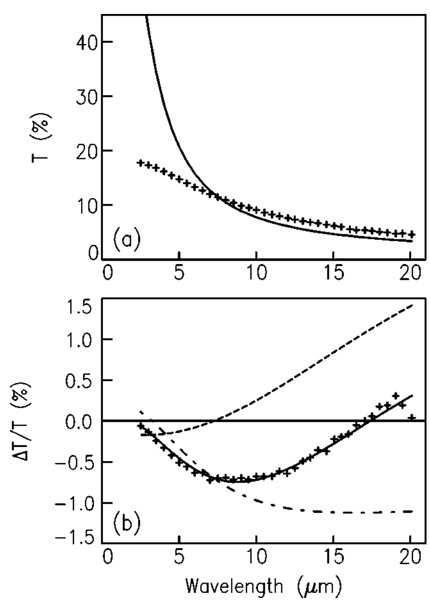

FIG. 3. Experimental data for transmission (a) and linear dichroism effect (b) of 19 nm $Ni_{80}Fe_{20}$ (+) together with the fit using the model discussed in the text and the model parameters as given in Table I (full curve). Also given in the lower graph is the calculated linear dichroism effect assuming either $a^{\downarrow}=0$ (dashed curve) or $a^{\uparrow}=0$ (dashed-dotted curve), all other model parameters remaining the same.

mains approximately constant at 2.5±0.2, $a^{\uparrow}$ increases from 0.031 to 0.067, and $a^{\downarrow}$ becomes less negative, changing from −0.060 to −0.015. The fits also show that the smallest values of $n^{\downarrow}/m^{\downarrow}$ give the best fit of the transmission as a function of wavelength.

This shows that the analysis of the magnetic linear dichroism effect provides a sensitive new method for studying spin-polarized electron transport in metal films. Within the model used, electron transport in all films studied is to be considered as spin dependent at room temperature. At present no direct quantitative comparison can be made with results reported elsewhere. For $Ni_{80}Fe_{20}$ the analysis of the CIP-GMR effect of spin valves at room temperature has yielded $\lambda^{\uparrow}/\lambda^{\downarrow} \gtrsim 7.7$ (Ref. 7) (using a rather indirect method for obtaining $\lambda^{\downarrow}$). The analysis of the CPP-GMR effect at 4.2 K has yielded $\rho^{\downarrow}/\rho^{\uparrow} \approx 7.5 \pm ^{1.5}_{2.6}$, when taking spin-flip scattering into account as well.[13] The value of $\alpha = \tau_o^{\uparrow}/\tau_o^{\downarrow}$ as obtained for 19 nm $Ni_{80}Fe_{20}$ from our study at room temperature is significantly smaller. However, when calculating the mean free path ($\lambda = v_F \tau$), a possibly spin-dependent Fermi velocity has to be included.

In order to investigate the origin of the disagreement at small wavelengths between the calculated and experimental transmission, refinements of the model would be of much interest, e.g. including the effect of multiple internal reflections of the infrared light in the layers, the decay of the infrared light intensity in the layers, and spin-flip scattering.[14] Experimentally, interesting extensions would be the performance of *in situ* experiments (for which the films need not be covered with Ta protective layers), temperature dependent studies, studies of the effect in the reflection mode, the performance of spatially resolved studies, and the extension to larger wavelengths. Preliminary experiments for wavelengths up to 40 μm showed for $Ni_{80}Fe_{20}$ a further increase of $\Delta T/T$, although even at a wavelength of 40 μm the expected high wavelength limiting value ($\Delta T/T = \Delta\rho/\rho$) was not yet reached.

In conclusion, we have found a linear magnetic dichroism effect for infrared light in ferromagnetic alloys. It has been found that the amount of infrared light transmitted through $Ni_{80}Fe_{20}$, $Ni_{80}Co_{20}$, $Co_{90}Fe_{10}$, and $Fe_{88}V_{12}$ thin films depends on the angle between the polarization direction of the light and the magnetization direction of the film. The linear dichroism effect has been analyzed in terms of a two-current Drude-type model for the (frequency dependent) conductivity. Analysis of the measurements with this model produces the spin- and angular-dependent relaxation times.

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