Infrared spectra of giant magnetoresistance Fe/Cr/Fe trilayers

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Magnetic-field-induced changes in infrared transmission and reflection from Fe/Cr/Fe trilayers are reported. Changes as large as ≈1% (compared with 4–5 % changes in resistivity) are observed around 2000 cm^{-1}, and the magnitude of the effect decreases monotonically to zero at ≈5000 cm^{-1}. The field dependence mimics that of the resistivity, and saturates at the same field at which the magnetization of the two Fe layers align parallel to each other. A simple model, which estimates the frequency dependence of the resistivity and includes the frequency dependence of the skin depth, produces semiquantitative agreement with experiment. [S0163-1829(98)02206-1]

The giant magnetoresistance (GMR) observed in many ferromagnetic/nonmagnetic metallic multilayer systems[1,2] is a phenomenon which has attracted considerable attention both because of its potential for applications as well as its intriguing origin related to the spin polarization of the non-magnetic layers. The surface magneto-optical Kerr effect (SMOKE)[3] has been used with considerable success to investigate these materials. The technique yields detailed information on the spin orientation of the constituent layers and is well understood in terms of bulk magneto-optic constants. It appears therefore that the optical properties of GMR materials are well understood in the visible range. Contrary to this situation, there is little known about the infrared (IR) properties of these materials. *A priori*, if the IR effects were comparable in magnitude to those in the visible (typically changes of one part in 10^5), they would not be observable. This is because conventional IR techniques lack intensity, stability, and polarization features which make visible SMOKE effects possible. We are aware of only two investigations of the IR response of magnetic multilayers.[4,5] The results in Ref. 4, on the Co/Cu/permalloy system, indicate that the magnetic-field-induced changes in the IR are considerably larger than those in the visible.

Here we present the results of an IR transmission and reflection study of antiferromagnetically (AF) coupled Fe/Cr/Fe trilayers. We observe large (≈1%) changes in the transmission as fields that align the two magnetic layers are applied. We develop a model to estimate the frequency dependence of the magnetoresistance. The model, which contains no freely adjustable parameters, reproduces both the order of magnitude and the frequency response of our observations.

The (211)- and (100)-oriented Fe/Cr/Fe trilayer samples were made by dc magnetron sputtering onto epitaxially polished single-crystal MgO (110) and (100) substrates, respectively, using the same growth procedure outlined for superlattices.[6] The 20-Å Cr buffer layers were grown at 600 °C. The substrates were then cooled to ≈150 °C prior to the growth AF-coupled FE(20 Å)/Cr(12 Å)/Fe(20 Å) trilayer which were then capped with a 20-Å Cr layer. The magnetic characterization of similarly grown Fe/Cr/Fe trilayers is described in Ref. 7.

Shown in Fig. 1(a) is the magnetoresistance measured on the (211)-oriented sample with H applied along the in-plane easy axis. We obtain a GMR value of 3.8% [for the (100)-oriented sample we obtain 5%]. The shape of the MR curve is characteristic of AF-coupled films with uniaxial anisotropy.[7] To measure the field-induced IR response, the samples were placed between the poles of an electromagnet and the transmission and reflectivity were measured as a function of the applied field H. Shown in Fig. 1(b) is the field-induced change (measured relative to the aligned state) in IR transmission measured at 2000 cm^{-1} for the sample shown in Fig. 1(a). The field-induced IR response mimics the MR showing that the two phenomena have a common origin. In particular, both the MR and $\Delta T/T$ saturate above 3.5 kG when the Fe layers become aligned parallel to each other. However, the IR response has the opposite sign and reduced magnitude as compared to the resistivity measurements.

To further explore this phenomena, we have studied its frequency dependence. A typical (Fourier transform IR) (FTIR) transmission (T) spectrum, measured in the mid-IR region and normalized to an MgO substrate, is shown in Fig.

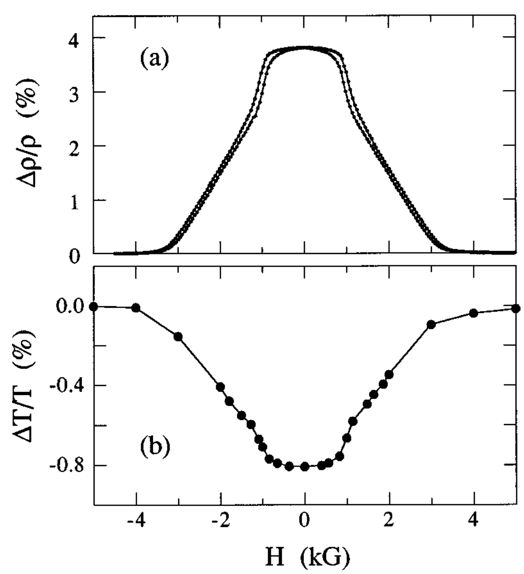

FIG. 1. The magnetoresistance (a), and field dependence of IR transmission at 2000 cm^{-1} (b), of the [112]-oriented sample for H along the easy axis.

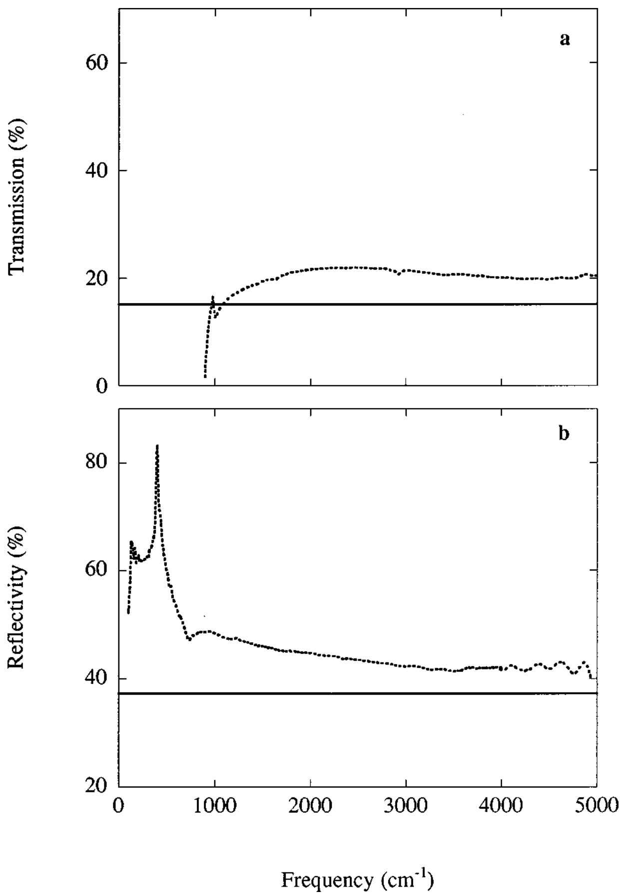

FIG. 2. (a) Transmission spectrum (dashed line) of our Fe/Cr/Fe trilayer, measured in the mid-IR region and normalized to an MgO substrate. (b) Reflectivity spectrum (dashed line) of the same sample normalized to that of a thick Cr film. The full lines are the fit to the data using Eqs. (2) and (3) with only $\mu\sigma$ as a free parameter.

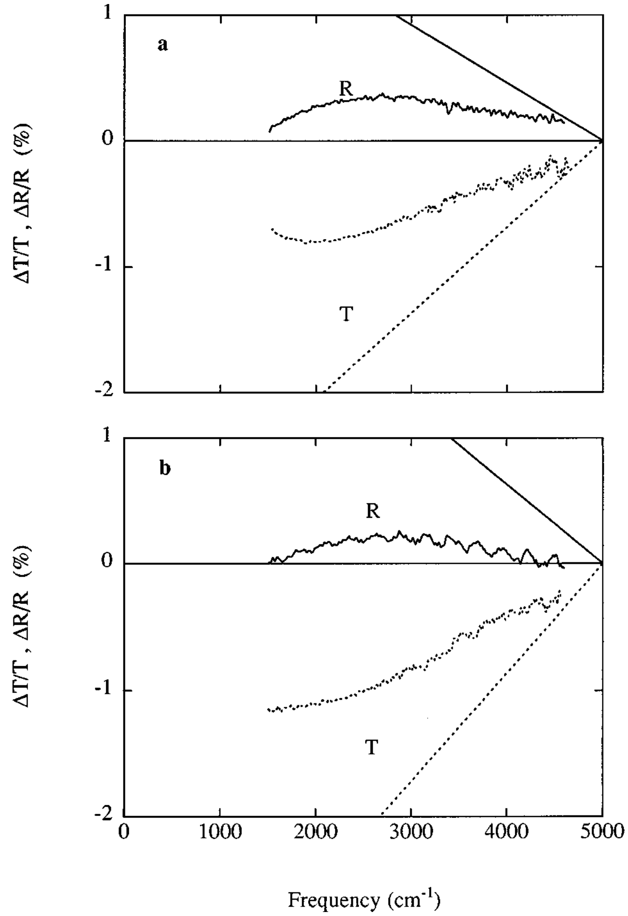

FIG. 3. Magnetic-field-dependent transmission (dashed) and reflectivity (full) spectra normalized to their high-field values. The straight lines are the results obtained from the model. (a) and (b) are for the (211) and (100) samples, respectively.

2(a). Below 1200 cm^{-1} the MgO substrate is either completely opaque or shows signs of magnetically induced transmission changes perhaps due to magnetic impurities. We therefore restrict our study to the ≈ 1200 to 5000 cm^{-1} range. The reflectivity (R), shown in Fig. 2(b), was measured in the mid- and far-IR regions and was normalized to that of a thick Cr film. Below 1000 cm^{-1} the reststrahlen from the MgO substrate dominates the reflectivity and clearly makes the data unsuitable for analysis. Figures 3(a) and 3(b) show the frequency dependence of the field-induced IR response $\{(T_{H=0}-T_{\mathrm{sat}})/T_{\mathrm{sat}}$ and $(R_{H=0}-R_{\mathrm{sat}})/R_{\mathrm{sat}}\}$ for the (211) and (100) samples, respectively, and H along the easy axis. At any given frequency, the field dependence is analogous to that observed in Fig. 1(b) but with the magnitude of the effect varying with frequency. The IR response is a maximum at ≈ 2000 cm^{-1} and decays monotonically with increasing frequency to zero at ≈ 5000 cm^{-1}.

To confirm that the phenomena exhibited in Fig. 3 are indeed due to the Fe/Cr/Fe trilayer structure, we also performed identical measurements on pure MgO substrates and on an MgO substrate on which a single 40-Å Fe layer (equal in thickness to the two Fe layers of the trilayer) and with identical buffer and capping Cr layers. The MgO substrate showed a large and unexplained sharp decrease in transmission at around 1000 cm^{-1} as a field was applied. It is possible that a small portion of the slope change in Figs. 3 and 4 could be due to the tail of this feature. The sample with the single Fe layer behaved in a similar manner to the MgO substrate, but neither showed any field-induced changes in transmission over the range 1500–4000 cm^{-1}.

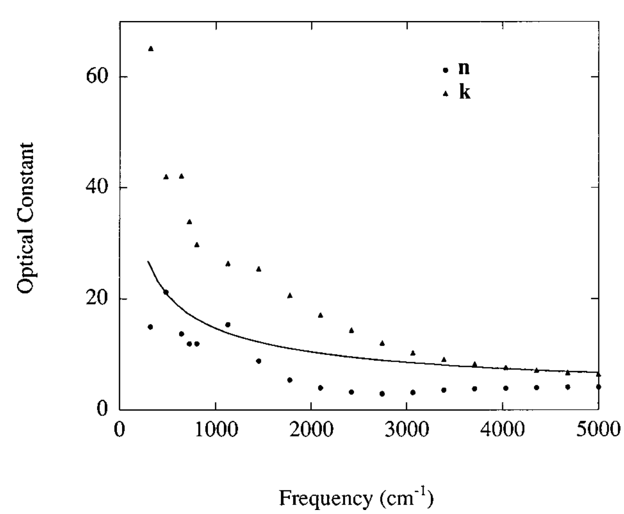

FIG. 4. Fitted index of refraction n $(=\kappa)$ is plotted as the full line. The literature values (Ref. 9) for bulk Cr are also shown.

Following the theory for electromagnetic waves in conducting media,[8] it can be shown that for good conductors the real (n) and imaginary (κ) parts of the refractive index are given by

$$n = \kappa = c/\omega\delta \quad \text{and} \quad \delta = (2/\omega\mu\sigma)^{1/2}, \tag{1}$$

where δ is the skin depth, c is the velocity of light, μ the permeability, ω the frequency, and σ the conductivity. The transmission and reflectivity of a metal film on a substrate are readily calculated in terms of n $(=\kappa)$. Following Ref. 8 we have

$$\begin{aligned} T = \{ & [(1-R_{12})(1-R_{23}) \\ & -4(R_{12}R_{23})^{1/2}\sin\delta_{12}\sin\delta_{23}]e^{-2(n\omega d/c)}\}/\{n_0[1 \\ & +2(R_{12}R_{23})^{1/2}e^{-2(n\omega d/c)}\cos(\delta_{12}+\delta_{23}+2n\omega d/c) \\ & +R_{12}R_{23}e^{-4(n\omega d/c)}]\} \end{aligned} \tag{2}$$

and

$$\begin{aligned} R = [& R_{12}+2(R_{12}R_{23})^{1/2}e^{-2(n\omega d/c)}\cos(\delta_{23}-\delta_{12}+2n\omega d/c) \\ & +R_{23}e^{-4(n\omega d/c)}]/[1+2(R_{12}R_{23})^{1/2}e^{-2(n\omega d/c)} \\ & \times\cos(\delta_{12}+\delta_{23}+2n\omega d/c)+R_{12}R_{23}e^{-4(n\omega d/c)}], \end{aligned} \tag{3}$$

where n_0 is the refractive index of MgO, R_{12} and R_{23} are the reflection coefficients at normal incidence for a plane interface dividing two semi-infinite media, and δ_{12} and δ_{23} are the corresponding phase angles. They are given by

$$\begin{aligned} R_{12} &= [(1-n)^2+n^2]/[(1+n)^2+n^2], \\ R_{23} &= [(n-n_0)^2+n^2]/[(n+n_0)^2+n^2], \end{aligned} \tag{4}$$

and

$$\begin{aligned} \delta_{12} &= \arctan[-2n/(1-2n^2)], \\ \delta_{23} &= \arctan[2nn_0/(2n^2-n_0^2)]. \end{aligned} \tag{5}$$

Using Eqs. (1)–(5) we have adjusted $\mu\sigma$ to fit our data in Figs. 2(a) and 2(b); the full lines are the results of such a fitting procedure. The agreement between the fit and the data is reasonable, especially given the simplicity of the model (i.e., only the product of $\mu\sigma$ is an adjustable parameter). In Fig. 4 we plot (full line) our fitted value of n $(=\kappa)$ and compare it to the values for bulk Cr.[9] The agreement again confirms that the approximations being made are realistic. We mention also that the same calculation using a wavelength dependent n for the MgO substrate [1.29 at 1000 cm^{-1} to 1.71 at 5000 cm^{-1} (Ref. 9)] does not produce any significant differences.

Our first attempt to fit the data in Fig. 3 was to change the single free parameter discussed above. Such an approach explains both the sign of the IR response and the close relation between the IR and MR results. However, changing the conductivity by $\approx 4\%$ as suggested by the magnetoresistance data, leads to a frequency independent change, also of about $\pm 4\%$, in T and R. This prediction is clearly inadequate to account for the observed magnitude and frequency dependence of the experimental results.

Within the framework of our simple model the origin of the observed frequency dependence can only originate in the conductivity. Guided by this, we note that since the magnetoresistivity is proportional to the number of electrons that probe *both* Fe/Cr interfaces,[1,2] it is reasonable to assume that only those electrons that reach both interfaces within a period of the electromagnetic radiation $(2\pi/\omega)$ will contribute to the magnetoresistance. Since the travel time τ across the Cr film cannot be less than d/v_F, where v_F is the Fermi velocity, this immediately leads to a cutoff frequency ω_c. Using a typical Fermi velocity (1 Å/s) for a ''free-electron'' metal, and $d = 12$ Å, leads to $\omega_c = 30\,000$ cm^{-1}. The Fermi velocity for the electrons believed to be responsible for transmitting the spin information in Cr (viz., at either the lens or ellipse in the Brillouin zone) is roughly 6 times smaller[10] and consequently leads to a cutoff ≈ 5000 cm^{-1}. Since transport is more likely to result from the ellipse which has s-p character, a spherical Fermi surface appears to be a reasonable approximation. In this approximation only those electrons propagating at an angle θ from the surface normal such that

$$v_F\cos\theta/d > \omega/2\pi \tag{6}$$

will contribute to the magnetoresistance. As ω increases, the fraction of electrons that can probe both interfaces decreases and, hence, the magnitude of the effect decreases. For $\omega > \omega_c = 2\pi v_F/d$ no electrons can reach both interfaces within $2\pi/\omega$ and, thus, the magnetoresistive effect disappears. For a spherical Fermi surface it is trivial to integrate Eq. (6); it leads to

$$\begin{aligned} \Delta\sigma/\sigma &= (\Delta\sigma/\sigma)_0(1-\omega/\omega_c) \quad \text{for } \omega < \omega_c, \\ \Delta\sigma/\sigma &= 0 \quad \text{for } \omega > \omega_c. \end{aligned} \tag{7}$$

Using Eqs. (1)–(7) and $\Delta\sigma/\sigma_{(\omega=0)}$ 3.8% from Fig. 1(a), we obtain the dashed lines shown in Fig. 3. Given the simplicity of the model we feel that there is satisfactory agreement between the experimental results and the model.

The experimental results and the model presented above must also be viewed in the context of the results and the model presented in Ref. 4. Experimentally there is agreement in that large, field-induced effects are observed. There are, however, some puzzling differences in detail which we are unable to reconcile. In Ref. 4 the transmission (in zero field) increases with frequency—compared to our results (Fig. 2) which are essentially flat when substrate effects are not present. The origin of the wavelength dependence in Ref. 4 is not clear, especially since at even shorter wavelengths (i.e., visible light) their $\approx$250-Å films should become opaque. The other feature which we do not understand is the reported change in sign of $\Delta T/T$ (negative to positive as the wavelength increases) as shown in Fig. 2 of Ref. 4. Using Eqs. (2) and (3) we are unable to obtain a sign change without changing the sign of $\Delta\sigma$ which would appear to be unphysical. The sign reversal is also puzzling because at zero frequency one might expect the sign to be that determined by the dc resistivity (viz., negative), and hence, a second sign change would be required in the model of Ref. 4 at smaller frequencies than those measured. However, it should be kept in mind that the model developed in Ref. 4 produces agreement with their experimental results. The model in Ref. 4 is also more advanced than our present model since it contains a nonlocal description of the dielectric constant. Unfortunately, the details of the fitting procedure used in Ref. 4 (their Ref. 25) has eluded us and hence we are unable to make a quantitative comparison.

Kerr measurements on similar samples[7] may appear to contradict the model presented here which predicts that all magnetooptic effects above 5000 cm^{-1} should vanish. However, the origin of the weak magnetooptic effects in the visible region in Ref. 7 can be traced to the individual Fe layers. As mentioned in the introduction the magnitude of the magneto-optic effects in pure Fe are orders of magnitude smaller than those presented in Figs. 1 and 3 and are not detectable on the scale of the IR data. They only become observable in the visible due to the existence of high performance optics, lasers and detectors.

In conclusion, we have observed large, magnetic-field-induced changes in IR transmission and reflectivity in Cr/Fe/Cr trilayers. A model is presented which semiquantitatively accounts for the observed phenomena, including the magnitude of the changes, the scaling with field, and the frequency dependence. There are also some shortcomings of the model, and of the experiments themselves, which require further investigation. For example, the use of MgO substrates complicates the analysis due to its strong absorption and reststrahlen, although the MgO facilitated the growth of single crystal films which simplifies part of the data interpretation. Experiments on Si substrates might improve the low frequency data. From the theoretical standpoint it is necessary to reformulate the simplistic arguments suggested here into a rigorous theoretical framework. Such an approach must also include the realistic band structure of the spacer layer.

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