

Low Frequency Magnetic Response in Antiferromagnetically Coupled Fe/Cr Multilayers

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The magnetic field and temperature dependence of the low frequency magnetic response of antiferromagnetically coupled Fe/Cr(100) multilayers has been studied between ± 500 Oe, from 2 to 300 K. At $T = 2$ K the losses exhibit an unusually strong frequency dependence which can be described within a single relaxation time scheme. This relaxation time proves to be strongly field dependent. These phenomena are specific for epitaxial multilayers with large magnetoresistance. The behavior of the relaxation time at low temperatures might be related to some quantum tunneling processes.

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The study of dynamical properties of antiferromagnetically coupled magnetic multilayers (MML) is an important subject both from the fundamental and the applied point of view. A great number of available data are related to the high frequency (GHz) range [1–3]. This is quite natural because the authors were interested in the collective spin wave modes of the superlattices. When treating the spin-wave spectrum, one considers that the magnetization within a layer is homogeneous. This, however, is far from being the case. The interface roughness, as well as other factors, leads to magnetic inhomogeneities within every layer. The resulting structure is usually referred to as a domain structure although it is not very consistent, because the characteristic scales of the inhomogeneities are smaller than the domain wall widths [4]. Response of this structure to the ac fields has little to do with the spin waves. It treats, rather, processes associated with overcoming some barriers, pinning, depinning, etc. The corresponding dynamics clearly belongs to a low frequency range, which remains practically unexplored up until now.

Indeed, the only data on the low frequency response of MML are those of Ref. [5], where the real (χ') and the imaginary (χ'') parts of the magnetic susceptibility of a superlattice Fe/Cr(211) were measured at a frequency $f = 10^3$ Hz and at a nonspecified temperature. Along with maxima almost coinciding with those of $\chi'(H)$ (and associated with spin-flop transitions discussed in the paper) $\chi''(H)$ had an additional maximum at $H = 0$. This maximum had clearly no relevance to the spin-flop transitions but reflected some properties of the domain structure. We think that already the data of Ref. [5] provide evidence that the low frequency response of MML bears nontrivial (albeit not easily interpretable) information about the domain structure and deserves a systematic study.

In this Letter we report on a detailed study of the ac magnetic susceptibility of antiferromagnetically coupled Fe/Cr(100) multilayers at different frequencies below 10^4 Hz, magnetic fields below 100 Oe (with full sweep between ± 500 Oe), and at temperatures down to 2 K. In

these multilayers (for a recent review see [6]) the presence of the antiferromagnetic exchange [1] has led to the discovery of the giant magnetoresistance (GMR) [7] and an oscillatory exchange coupling [8]. We measured both the real χ' and the imaginary χ'' parts of the magnetic susceptibility defined as $M(\omega)/H(\omega)$ [here $H(\omega)$ is an ac driving field (acdf) and $M(\omega)$ is the corresponding magnetic response].

The epitaxial $[\text{Fe/Cr}]_n$ ($n = 10, 40,$ and 50) multilayers on MgO(001) and polycrystalline $[\text{Fe/Cr}]_{10}$ on yttria stabilized zirconia (YSZ) substrates were deposited in a molecular beam epitaxy system held at a temperature of 50°C [9]. The magnetic properties have been studied using the *Magnetic* ($3\text{ Hz} < f < 167\text{ Hz}$) and *Physical* ($77\text{ Hz} < f < 9876\text{ Hz}$) Property Measurement Systems (*Quantum Design*) with static and alternating magnetic fields parallel to each other and to the film's surface. Magnetization measurements showed that, for epitaxial $[\text{Fe/Cr}]_{10}$ multilayers deposited on MgO(001), the GMR at 10 K is about 100% and the antiferromagnetic fraction (AFF), defined as $\text{AFF} = 1 - (M_r/M_S)$, where M_r and M_S are, respectively, the remanent and saturation magnetization (above saturation field $H_S \sim 10\text{ kOe}$), 0.85 ± 0.05 . This deviation from 100% antiferromagnetic coupling is a characteristic feature of the Fe/Cr multilayers and is usually attributed to biquadratic coupling [10]. All data reported here, except those specified, have been reproduced for various epitaxial antiferromagnetically coupled $[\text{Fe/Cr}]_{10}$ MML with GMR above 100%.

The left inset in Fig. 1a shows low field ($H < 0.2H_S$) magnetization for $[\text{Fe}(30\text{ \AA}/\text{Cr}(13\text{ \AA}))]_{10}$ MML at 10 K and magnetic field along (110). The upper right inset clearly proves that low field hysteresis ($H < 100$ Oe) is not due to easy/hard axis orientation transitions in four-fold magnetic symmetry, which is reflected in weak maxima at about 300 Oe. The main part in Fig. 1 expands the low field dependence of the real (Fig. 1a) and imaginary (Fig. 1b, in semilogarithmic scale) contributions of the magnetic susceptibility in $[\text{Fe}(30\text{ \AA}/\text{Cr}(13\text{ \AA}))]_{10}$. At high

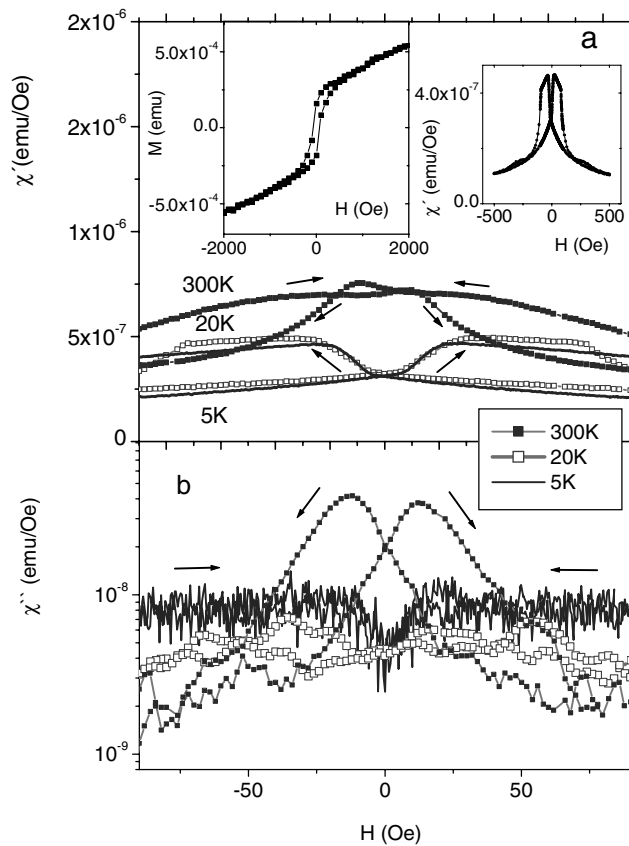


FIG. 1. Real (a) and imaginary (b) contributions to the magnetic susceptibility of a $[\text{Fe}(30 \text{ \AA})/\text{Cr}(13 \text{ \AA})]_{10}$ multilayer measured at 987 Hz with $\text{acdf} = 4$ Oe and at temperatures 300, 20, and 5 K. The data (b) for $T = 20$ and 300 K have been averaged (over 5 points). The upper left inset in (a) shows the low field part of the magnetization vs magnetic field at 10 K. The right inset shows the real part of the magnetic susceptibility for $H < 500$ Oe. Orientation of the magnetic field is along (110).

temperatures ($T > 100$ K), independently of the applied acdf (2–8 Oe), both $\chi'(H)$ and $\chi''(H)$ show hysteretic dependence on the magnetic field with the maxima of the losses at $H \approx \pm(20-30)$ Oe. The observed maxima as the field is swept from +500 Oe down to zero and then to negative values (or in the opposite direction) can be naturally explained by the formation of small-scale magnetic structures repeatedly observed in MML. The presence of the domain structure for the same sample was demonstrated by using magnetic force microscopy [11].

More interesting is the dependence of the losses on temperature. We refer here to the losses in low magnetic fields ($H \approx 50$ Oe). Lowering the temperature from 300 to about 10 K, the losses somewhat decrease. At lower temperatures the losses begin to *increase*. The losses at 5 K are *higher* than at 300 K (beyond the maxima observed at the latter temperature). At 2 K they are even higher. The character of the dependence of the losses on the magnetic field changes as well. If the acdf does not exceed some critical value (≤ 4 Oe), the hysteretic dependence of the losses on the magnetic field gradually disappears be-

low approximately 50 K. One more remarkable feature is an appearance at lower temperatures ($T < 7$ K) of a minimum in the magnetic losses at $H = 0$ (see Fig. 1b).

More details about the dependence of χ'' on H and temperature are presented in Fig. 2. One sees that the lossy part of the susceptibility is higher at 2 K than at 5 K or 7 K, and the narrow (with half width of about 15 Oe) zero field minimum in magnetic losses becomes more pronounced. We have not detected essential changes in this dip measured for magnetic fields directed along either the easy or hard axes. The imaginary part of the magnetic susceptibility of the MgO(100) substrate at 2 K and $H < 100$ Oe is smaller than 2×10^{-9} emu/Oe and shows no signs of a narrow dip for $H < 15$ Oe. This means that the observed effect is not extrinsic.

The frequency dependence of χ'' proves to be very surprising. We studied it at $T = 2$ K and $T = 10$ K. At higher temperatures the out-of-phase susceptibility is too small to investigate its frequency dependence. The main surprise is that it exhibits a well-pronounced frequency dependence at $T = 2$ K ($H = 50$ Oe) and this dependence may be reasonably fitted by the single relaxation time formula $\chi'' = \chi_0 \frac{\omega\tau}{1+(\omega\tau)^2}$ with $\tau \approx 2.5 \times 10^{-4}$ s (see inset in Fig. 3). One normally expects the response of domain structures to be characterized by a broad distribution of the relaxation times with χ'' almost independent of frequency. This seems to be the case for higher temperatures: the frequency dependence of χ'' at $T = 10$ K is nearly non-existent with a much broader maximum shifted to higher frequencies. Note that in the frequency range studied the real part of the susceptibility is, within a margin of 20%, independent of f between 3 and 9876 Hz. The fact that

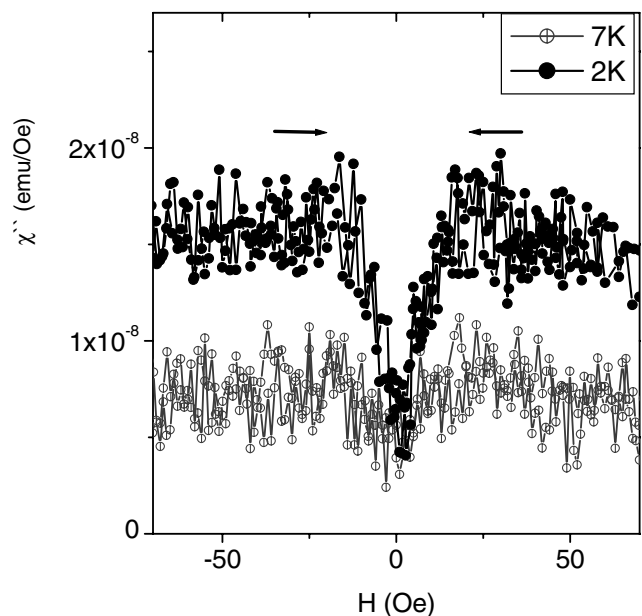


FIG. 2. Imaginary part of the magnetic susceptibility of $[\text{Fe}(30 \text{ \AA})/\text{Cr}(13 \text{ \AA})]_{10}$ multilayer measured at 987 Hz with $\text{acdf} = 4$ Oe and at temperatures 2 and 7 K.

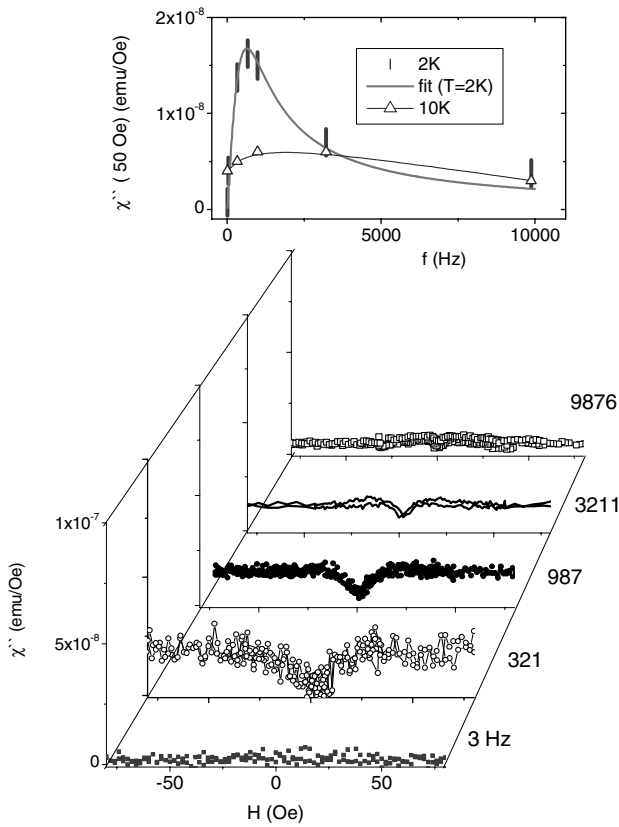


FIG. 3. Imaginary part of the magnetic susceptibility of $[\text{Fe}(30 \text{ \AA})/\text{Cr}(13 \text{ \AA})]_{10}$ multilayer measured at five different frequencies with $\text{acdf} = 4 \text{ Oe}$ at 2 K. The upper graphic shows the frequency dependence of the dissipation at small nonzero magnetic field ($H = 50 \text{ Oe}$) at temperature $T = 2$ and 10 K. The bold solid line represents the fit described in the main text.

the real part is not described within the one relaxational mode model is not surprising: this mode may be responsible only for a part of the magnetic response although it governs the low frequency losses.

Quite spectacular is the dependence of χ'' on the magnetic field. Figure 3 shows $\chi''(H)$ measured at five different frequencies between 3 and 9876 Hz. One sees nontrivial behavior of χ'' at low fields: a well-defined minimum in $\chi''(H)$ at $H = 0$ for $f = 321$ and 987 Hz, a more narrow minimum flanked by two symmetric maxima for $f = 3211$ Hz, and similar behavior but less pronounced and with an even narrower minimum for $f = 9876$ Hz. All these curves can be qualitatively explained if one assumes that the above relaxation time decreases as H diminishes and $\tau(H = 0)$ is less than $\tau(H = 50 \text{ Oe})$ by about an order of magnitude. Indeed, as τ decreases the maximum of the curve in the inset of Fig. 3 shifts to the right. One sees that the points corresponding to $f = 321$ and 987 Hz move away from the maximum. The point corresponding to $f = 3211$ Hz first approaches the maximum (and χ'' increases) and then moves away from the maximum (and χ'' decreases). The same happens with the point corresponding to $f = 9876$ Hz but, of course, it

passes through the maximum at a smaller field. We see that the model of a single relaxation time allows us to explain the dependence $\chi''(f, H)$ as well.

In order to investigate the relationship between the observed effects and the degree of antiferromagnetic coupling, we measured $\chi'(H)$ and $\chi''(H)$ for the samples with different GMR between 16% and 220% (Fig. 4). While the Fe/Cr sample with a record magnetoresistance (220%) [9] among MMLs shows a behavior similar to that presented in Figs. 1–3 for samples with GMR about 100%, the magnetic field dependences of the losses for $[\text{Fe}/\text{Cr}]_{40}$ MML with GMR of about 60% show clearly both a weak nonhysteretic minimum in $\chi''(H)$ at about $H = 0$ and a hysteretic maximum at low fields. We note also that for epitaxial $[\text{Fe}(30 \text{ \AA})/\text{Cr}(16 \text{ \AA})]_{10}$ on $\text{MgO}(100)$ MML, with a Cr thickness well outside the maximum of antiferromagnetic coupling, the low field magnetic losses at 2 K (not shown here) could not be fitted by using a single relaxation time, and a wide minimum in $\chi''(H)$ at about $H = 0$ was observed only for $33 < f < 100 \text{ Hz}$. On the other hand, the low frequency magnetic response of weakly coupled Fe layers in nonepitaxial $[\text{Fe}/\text{Cr}]_{10}$ MML with

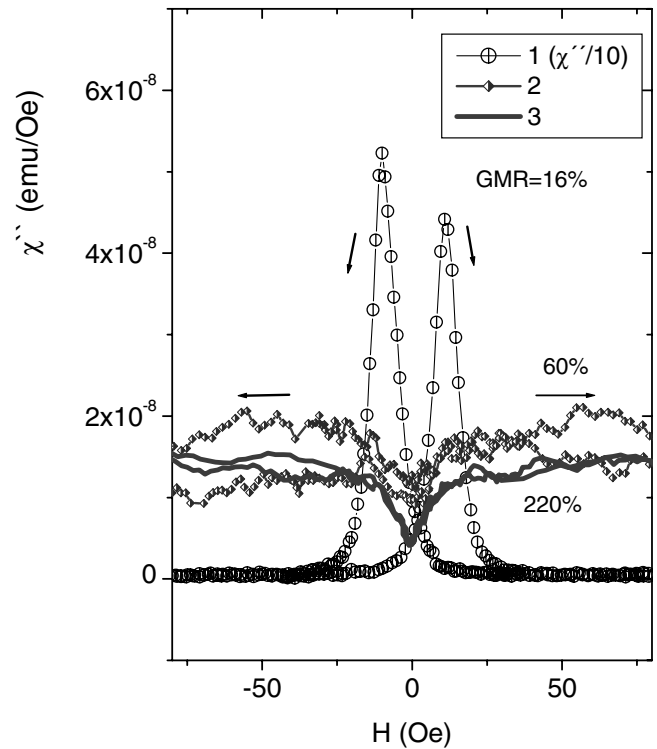


FIG. 4. Imaginary part of the magnetic susceptibility of three Fe/Cr MMLs with different degrees of antiferromagnetic coupling. Magnetic susceptibility was measured at temperature 2 K with $f = 987 \text{ Hz}$ and $\text{acdf} = 4 \text{ Oe}$. Curve 1 shows $\chi''(H)$ for the polycrystalline $[\text{Fe}(30 \text{ \AA})/\text{Cr}(13 \text{ \AA})]_{10}$ multilayer grown on YSZ. Curve 2 corresponds to $[\text{Fe}(15 \text{ \AA})/\text{Cr}(10 \text{ \AA})]_{40}$ grown on $\text{MgO}(001)$ and curve 3 shows $\chi''(H)$ for $[\text{Fe}(5 \text{ \AA})/\text{Cr}(12 \text{ \AA})]_{50}$ on $\text{MgO}(001)$. The magnetoresistance indicated near each curve was measured at liquid helium temperature. The curves 2 and 3 present averaged data (over 5 points).

enhanced interface roughness and small magnetoresistance is large for both real and imaginary (curve 1 in Fig. 4) parts and shows strong hysteresis in $\chi''(H)$ down to 2 K, but not the specific features we found in epitaxial antiferromagnetically coupled MMLs with GMR above 100%. Finally, we also point out that Fe(50 Å)/Au(20 Å) film did not show low temperature magnetic properties (presented in Figs. 1–3). Therefore, the observed phenomena are specific for multilayers with high GMR and strong antiferromagnetic coupling.

What could be the reason behind such unusual behavior of the losses at low temperatures? Part of the temperature evolution of the dependencies $\chi''(H)$ might be due to the evolution of the domain structure which remains basically unknown for low temperatures. However, the changes are too drastic to be accounted for by a variation of the domain structure alone. It is, of course, rather surprising that the one relaxation time model describes the important features of the *out of phase* response in a multidomain system. What is the meaning of these nearly identical relaxors? At present it is clearly premature to speculate about this. It is tempting to relate the decrease in the relaxation time to tunneling processes which provoke a “chain” or a “shock wave” of other processes, thus leading to a rapid relaxation. An enlightening discussion of such a possibility was given in Ref. [12].

It would be very interesting to study the low temperature and low frequency losses near $H = 0$ in other antiferromagnetically coupled MML's. The advantage of the Fe/Cr system is that the *epitaxial* superlattices can be grown on an isolating substrate (MgO) without buffer layers, diminishing drastically the losses due to the eddy currents. We were unable to detect these losses in our experiments. In some MMLs, such as Co/Cu, only growth on metallic [13] or insulating (but with a Nb buffer layer [14]) substrates provides good epitaxy. However, such MMLs exhibit losses due to the eddy currents of the buffer layer which may overshadow the losses of magnetic origin.

In conclusion, we have shown that the out of phase low frequency magnetic response in antiferromagnetically coupled Fe/Cr multilayers is strongly dependent on tem-

perature, magnetic field, and, at very low temperatures, on frequency. Magnetic losses at a nonzero (and fairly low) magnetic field first decrease but then *increase* with the lowering of the temperature. At temperatures below 7 K and for the ac drive frequencies $f \sim 10^2$ – 10^3 Hz we observed a dip in the magnetic field dependence of losses for fields $H < (10$ – $15)$ Oe. At $T = 2$ K and $H = 50$ Oe the frequency dependence of the losses can be satisfactorily described within a single relaxation time scheme. The dependence of χ'' on the magnetic field can be interpreted as the field dependence of the relaxation time which decreases by an order of magnitude as the field changes from $H = 50$ Oe to 0. The strong magnetic field dependence of the relaxation time at low temperatures might imply an involvement of quantum tunneling phenomena.

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