Temperature-independent magnetic relaxation in rare-earth layers

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(Received 14 February 1995; revised manuscript received 31 May 1995)

We report a study of temperature-independent magnetic relaxation in multilayers based on anisotropic rare earths R(Tb,Dy,$Dy_{50}Co_{50}$,Nd) of the form R/Mo. We find a temperature-independent magnetic relaxation below a certain temperature T_0 which depends on the measuring field and rare earth for all of the anisotropic rare-earth systems studied including those with nonferromagnetic order. Elemental Tb layers show the largest value of T_0 being 29 K and exhibit a sharp crossover at T_0. We consider a mesoscopic quantum tunneling (MQT) interpretation of this crossover. While there is a general trend for T_0 to increase with increasing anisotropy strength, quantitative comparison with a simple MQT model indicates there is significant disagreement between experiment and theory for the Tb system. We also find no significant change in T_0 when the Dy system is alloyed with Co. We consider other explanations of the temperature-independent magnetic relaxation and point out that a model in which interactions between domains walls are included may explain these results.

I. INTRODUCTION

In recent years magnetic relaxation studies at low temperatures have revealed a temperature-independent magnetic relaxation in a number of multilayer and fine particle systems based on transition metals or rare earths.[1–5] The crossover temperature T_0 below which the magnetic relaxation becomes temperature independent varies from system to system. In all cases in which detailed observations are reported these magnetic systems are complex and are known to have a distribution of energy barriers $N(E)$ (number of energy barriers with height E) associated with a pinning of the magnetization. The observed logarithmic form of the magnetic relaxation with time derives from this distribution of barriers.[6]

A great deal of attention has been given in the literature to a possible mesoscopic quantum tunneling mechanism as an explanation of this temperature independent relaxation.[1–5] In the magnetic systems studied thus far it is expected that if quantum tunneling occurs, it will involve 10^3–10^4 moments and so is referred to as mesoscopic quantum tunneling (MQT). If the observed temperature-dependent relaxation is associated with MQT then T_0 is the temperature at which the system crosses over to a regime where the magnetic relaxation is dominated by quantum tunneling through a barrier rather than thermal excitation over a barrier.[7–9] Support for a MQT interpretation comes from the measured value of T_0 which is found to be within the range expected from the estimated anisotropy and known magnetic moment of the systems studied so far.[1–4]

Two other mechanisms may also lead to a temperature-independent magnetization. In the first of these, it has been pointed out that since a distribution of barrier heights exist there is also the possibility that thermal activation along with an unusual distribution of barrier heights may lead to the observed plateau in $S(T)$.[10,11] The second mechanism involves a model in which magnetic domain walls in thin films interact. When one domain wall moves it may perturb a nearby domain wall causing it to relax to a more stable state. At low temperatures, once a particular domain wall has moved, many other domain walls may be influenced and the system would be dominated by the propagation of this interaction through the system. The idea of self-organized critical behavior depends on these ideas[12] and has been proposed as an explanation of the ubiquitous $1/f$ noise in a wide range of types of system including electrical systems. Such a model may yield a temperature independent relaxation at low temperatures but no calculations are available for magnetic systems yet.

Among the above models MQT has been given most consideration. However it is fair to say that the current experimental situation neither confirms that this temperature-independent relaxation is related to MQT nor does it eliminate MQT as a cause.

The purpose of this work is to examine the temperature-independent relaxation in some selected simple elemental systems with strong anisotropy where the magnetic anisotropy is known and determine whether there is any quantitative agreement or disagreement with a MQT interpretation. These systems are based on anisotropic rare-earth multilayers and are of the form $R(t$ nm)/Mo(18 nm) where R represents Tb, Dy, Nd, and $Dy_{50}Co_{50}$ each with at least five bilayers. The anisotropic rare-earth incorporates a strong single ion magnetic anisotropy within the rare-earth layer of these multilayers.[13] The $Dy_{50}Co_{50}$ layer in the last system is amorphous as we discuss below and the magnetic anisotropy direction varies at random from site to site. The elemental rare-earth layers of Tb, Dy, and Nd have a crystallite size of order 20 nm and can be thought of as magnetic systems with a coherent anisotropy within a crystallite and an anisotropy axis that varies at random from crystallite to crystallite. Magnetization reversal in these strong anisotropy systems is dominated by domain-wall movement.[2,14] Domain walls are in metastable positions and either tunnel through or are thermally excited over the potential barrier associated with the pinning site leading to a time dependence of magnetization. Possible sources for the range of energy barriers include variation in the orientation of crystallites within the layer and pinning of domain walls at interfaces as compared to the bulk.

We have chosen to study the Tb and Dy elemental systems because (i) their low-temperature magnetic order is very simple being ferromagnetic, (ii) their magnetic anisotropy is known[15] (at least in bulk form) and so any dependence of T_0 on anisotropy strength may be determined, and (iii) the magnetic anisotropy of these systems is among the largest of all elemental ferromagnets so that if quantum tunneling occurs, the value of T_0 will be large.[7–9] Mo is chosen as the spacer layer since Tb and Dy are not soluble to any significant extent in Mo minimizing any problems associated with chemical homogeneity.

We have included Nd/Mo and $Dy_{50}Co_{50}$/Mo multilayers in our study to determine what effect a more complex type of magnetic order would have on the low-temperature relaxation. The magnetic order within the layer is a complex helical state for Nd,[13] and is sperimagnetic in amorphous $Dy_{50}Co_{50}$.[16] In this latter state the Co moments are aligned and the Dy moments form a fan about a direction opposite that of the Co due to the strong random magnetic anisotropy. The fan is opposite the Co moment due to an antiferromagnetic exchange coupling between the Dy and Co moments.

The thickness of the Mo spacer layer, being 18 nm, is large enough that the rare-earth-based magnetic layers in these systems are magnetically isolated. We base this on the observation that in the metallic systems studied thus far, magnetic interactions through a nonmagnetic layer appear to become small for nonmagnetic layer thicknesses greater than 5 to 9 nm depending on the system studied.[17]

We have made a short report on the possibility of MQT in Tb and Dy multilayers with t of approximately 70 nm (Ref. 5) and we have also reported on the interface anisotropy in some of these multilayers with t in the range 1–10 nm.[18]

II. EXPERIMENTAL

The samples of this work were prepared by sputtering onto a Ta substrate. Large angle x-ray diffraction using Cu $K\alpha$ radiation show the expected Mo bcc structure and the rare-earth hexagonal polycrystalline structure of Tb, Dy, and Nd for the Tb/Mo, Nd/Mo, and Dy/Mo multilayers. We have estimated the rare-earth crystallite size in our crystalline multilayers using $b=0.9\lambda/B\cos\theta$ where B is the full width at half maximum of an x-ray peak at angle 2θ, and $\lambda=0.154\,06$ nm.[19] We find a grain size $b=20$ nm in multilayers with $t>30$ nm and this grain size decreases as t is reduced below 30 nm. In the case of the $Dy_{50}Co_{50}$/Mo multilayers, the DyCo layers are amorphous with only a broad maximum at 35° 2θ in x-ray-diffraction measurements. Figure 1 shows examples of Cu $K\alpha$ x-ray diffractograms for a Dy/Mo and a $Dy_{50}Co_{50}$/Mo multilayer.

We have done smaller angle ($10°>2\theta>2°$) x-ray studies in multilayers with small bilayer spacing ($t<5$ nm, Mo thickness<2 nm) to access the quality of the multilayers and we see small angle maxima associated with the multilayer periodicity for multilayers with bilayer spacing down to 0.8 nm and some examples of these have been reported by us previously.[5,18]

For magnetic studies multilayers were prepared with t in the range 9–720 nm and the Mo layer thickness was held constant at 18 nm. The magnetization of the multilayers was measured using a Quantum Design MPMS5 superconducting quantum interference device magnetometer. In all of our measurements on time dependence, the magnet is placed in the persistent mode disconnecting it from the power supply so eliminating noise contributions from the magnetic-field power supply.

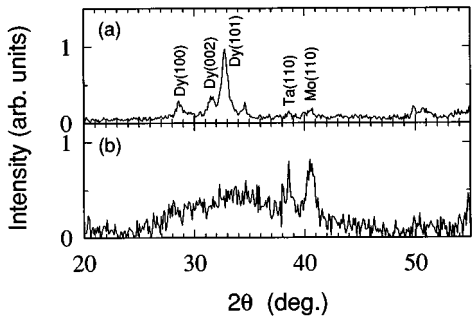

FIG. 1. X-ray diffractograms for (a) Dy(90 nm)/Mo(18 nm) and (b) $Dy_{50}Co_{50}$(81 nm)/Mo(18 nm) using Cu $K\alpha$ radiation. The peak indexes given are for hexagonal Dy and cubic Mo.

Figure 2 shows the zero-field-cooled (ZFC) and field-cooled (FC) magnetizations for a Tb and a Dy multilayer with the magnetic field applied in the plane of the sample. The initial rise in magnetization occurs at the expected bulk transition temperatures of 230 and 179 K for the Tb and Dy multilayers.[13] The second transition in Dy at 150 K is a transition from a helical magnetic state to the low-temperature ferromagnetic state. Both these systems have a large remanent magnetization at 4.5 K as expected for a ferromagnetic system as can be seen from the magnetization curves for selected multilayers in Fig. 3.

The strong magnetic anisotropy in these systems leads to

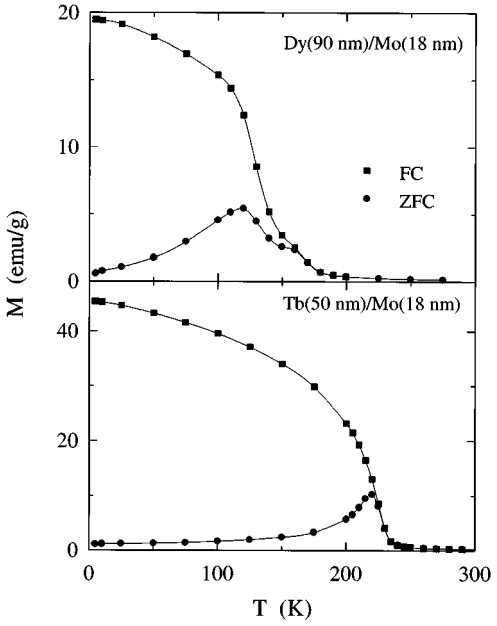

FIG. 2. Field-cooled (FC) and zero-field-cooled (ZFC) magnetizations for Tb and Dy multilayers in a field of 200 Oe. The solid lines are guides to the eye.

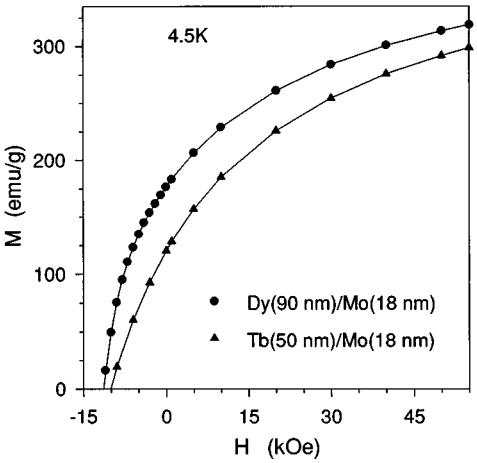

FIG. 3. Magnetization curves for selected Tb and Dy multilayers at 4.5 K. The solid lines are guides to the eye.

large magnetic hysteresis at low temperatures which manifests itself in a large difference between the FC and ZFC magnetizations as can be seen from Fig. 2. This strong anisotropy also leads to a large coercivity, greater than 10 kOe for the Tb and Dy multilayers as shown in Fig. 3.

Figure 4 shows examples of magnetic relaxation measurements at three selected temperatures for a Tb and a Dy multilayer. In this experiment the Tb multilayer is field cooled from 250 K (above the magnetic ordering temperature) in an applied field of 1 kOe to the measuring temperature, the applied field is then switched to the measuring value H_m, in this case -1 kOe. The time for the field reversal ranges from 65 to 85 s depending on the value of the final field. Reversing the applied field leaves the magnetization in a metastable state and the magnetization is then measured as a function of time. The Dy multilayer is field cooled from 200 K (again above the magnetic ordering temperature) in 100 Oe and then the field is reversed to its measuring value H_m, in this case -100 Oe. In the results described below both H_m and the measuring temperature are varied. As can be seen from Fig. 4, these data conform well to a logarithmic dependence of M on time of the form

$$M(t)=M_0[1-S(T)\ln(t/t_0)]. \tag{1}$$

Here $S(T)$ is the magnetic viscosity and is a measure of the relaxation rate and M_0 and t_0 are constants.

Figure 5 shows the variation of $S(T)$ calculated from Eq. (1) for a Tb/Mo multilayer for selected final fields. $S(T)$ becomes independent of temperature below a certain temperature T_0 which depends on H_m. The value of T_0 increases with decreasing H_m and reaches a maximum of 29 K at -200 Oe before decreasing below 20 K at 50 Oe. We see similar behavior in two other Tb multilayers.

We have performed similar experiments for the Dy multilayer and a plateau in $S(T)$ similar to that in our Tb multilayer is seen. Figure 6 shows these results as a function of H_m. Our largest T_0 for Dy is 6 K (± 0.5 K) at $H_m=-100$ Oe and is a factor of 5 smaller than Tb. A maximum is not seen in T_0 as a function of H_m but the low-field value does not appear to be increasing significantly with decreasing field. We have also gone to sufficiently high fields that the value of T_0 has been shifted below 4.5 K and a high field $S(T)$ curve is shown as an inset for Figs. 5 and 6 to illustrate this. We have summarized the dependence of T_0 on H_m for the Tb and Dy multilayers in Fig. 7.

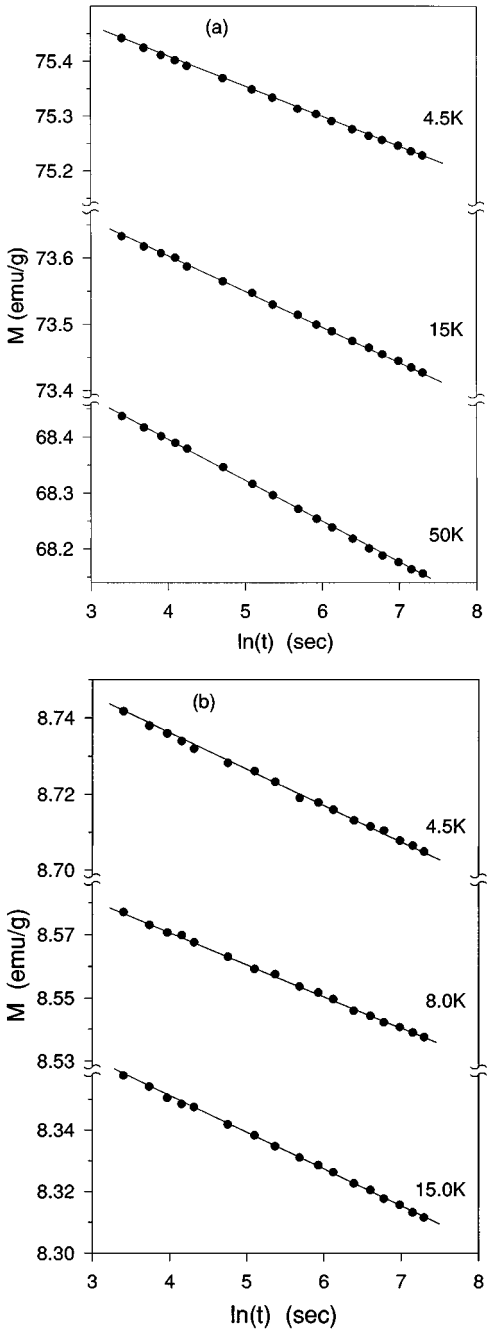

FIG. 4. Time dependence of magnetization for (a) the Tb(50 nm)/Mo(18 nm) and (b) Dy(90 nm)/Mo(18 nm) multilayers at three selected temperatures. The applied field and temperature were changed as described in the text. The solid lines are least-squares fits to Eq. (1).

To confirm that the observed temperature independent $S(T)$ at low temperatures is related to properties of thin films, we have looked at the time dependence of magnetization in bulk Tb and Dy. We find that at short times a logarithmic dependence of magnetization on time is present but we do not find that the coefficient $S(T)$ becomes temperature

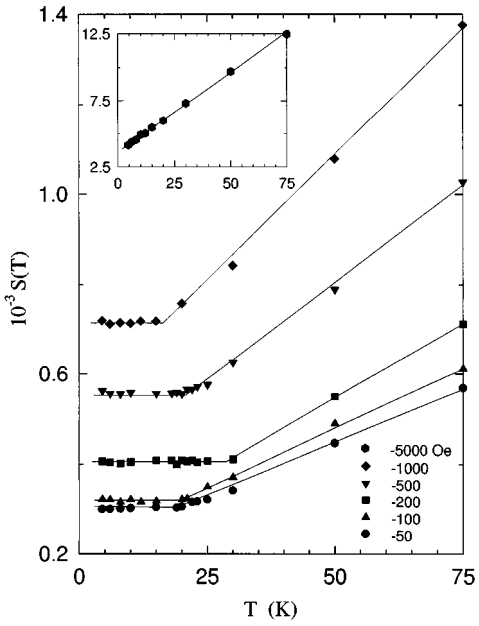

FIG. 5. Variation of the magnetic viscosity $S(T)$ [see Eq. (1)] with temperature for selected values of H_m, for the Tb(50 nm)/Mo(18 nm) multilayer. The unit of $S(T)$ is 1/ln(sec). The solid lines are guides to the eye.

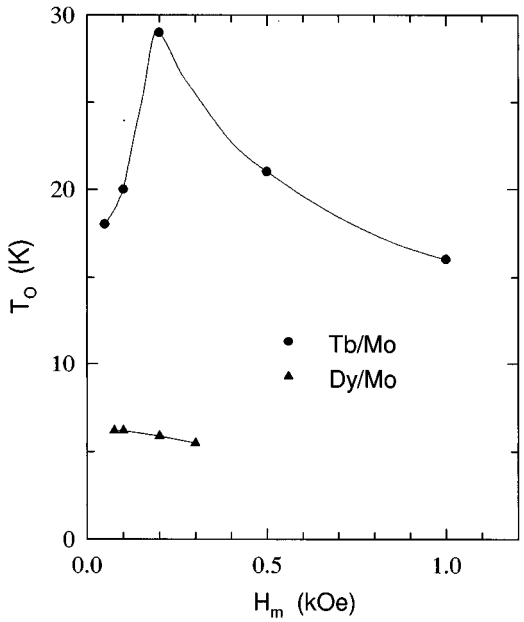

FIG. 7. Variation of the crossover temperature T_0 with measuring field H_m, for the Tb(50 nm)/Mo(18 nm) and Dy(90 nm)/Mo(18 nm) multilayers. The solid lines are guides to the eye.

independent at low temperatures. Rather, $S(T)$ shows a linear decrease with temperature down to 4.5 K for all our measuring fields.

In both the Nd(t nm)/Mo (18 nm) and $Dy_{50}Co_{50}$(t nm)/Mo (18 nm) where complex magnetic order is present, we see a temperature-independent magnetic relaxation at low temperatures. Figure 8 shows an example of the dependence of $S(T)$ on temperature for $H_m = -100$ Oe for a Nd multilayer and T_0 is approximately 10 K. The high-temperature behavior of $S(T)$ is not linear with temperature for this system. The highest T_0 for the DyCo multilayer system is 5.5 K (± 0.5 K) as can be seen for the $Dy_{50}Co_{50}$(650 nm)/Mo(18 nm) multilayer of Fig. 9. Thus there is only a small change in T_0 when Dy is alloyed with Co.

We have looked at the variation of T_0 with layer thickness for our multilayer systems and we find in all cases T_0 gradually decreases with increasing rare-earth layer thickness up to 720 nm, consistent with T_0 being below 4.5 K for bulk rare-

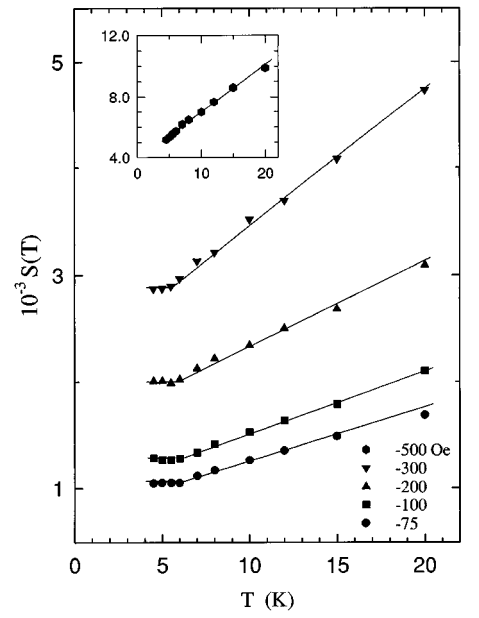

FIG. 6. Variation of the magnetic viscosity $S(T)$ [see Eq. (1)] with temperature for selected values of H_m, for the Dy(90 nm)/Mo(18 nm) multilayer. The unit of $S(T)$ is 1/ln(sec). The solid lines are guides to the eye.

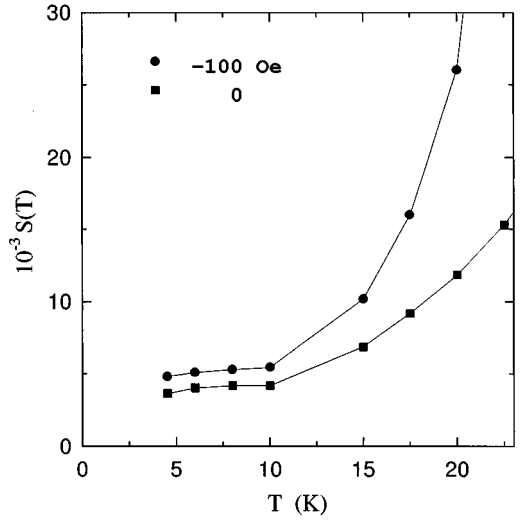

FIG. 8. Variation of the magnetic viscosity $S(T)$ [see Eq. (1)] with temperature for selected values of H_m, for the Nd(90 nm)/Mo(18 nm) multilayer. The unit of $S(T)$ is 1/ln(sec). The solid lines are guides to the eye.

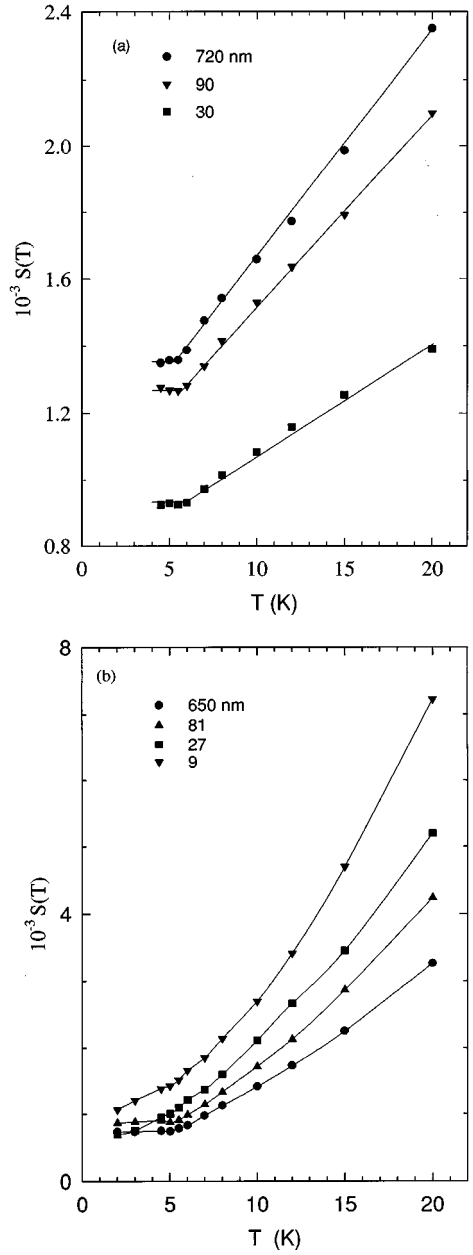

FIG. 9. Variation of the magnetic viscosity $S(T)$ [see Eq. (1)] with temperature with $H_m=-100$ Oe for selected rare-earth layer thickness t (given in the figure) for (a) Dy/Mo and (b) $Dy_{50}Co_{50}$/Mo multilayers. The unit of $S(T)$ is 1/ln(sec). The solid lines are guides to the eye.

earth systems. Examples are shown for the Dy and DyCo multilayers in Fig. 9 for H_m of -100 Oe.

III. DISCUSSION

We observe a temperature-independent magnetic relaxation at low temperatures for all of the anisotropic rare-earth multilayers studied including those that are not simple ferromagnets. At high temperatures (greater than T_0) the magnetic viscosity $S(T)$ is a linear function of temperature for the ferromagnetic systems of this work (Tb and Dy multilayers) and extrapolate to a nonzero value of $S(T)$ at $T=0$ K. Below a temperature T_0 which depends on the measuring field and the material $S(T)$ becomes independent of temperature so that the magnetic relaxation below T_0 is independent of temperature. We consider explanations for this behavior including the possibility of mesoscopic quantum tunneling, models involving various distributions of energy barriers $N(E)$, and the possibility of self-organized criticality.

We first consider the magnetic relaxation above T_0. For $T>T_0$ a linear dependence of $S(T)$ on temperature may be obtained in a model which assumes that $N(E)$ is constant over the energy range of barriers sampled by the magnetization. In this model $S(T)$ goes to zero as temperature goes to zero unlike our measurements where $S(T=0)$ is not equal to zero. A linear $S(T)$ that does not extrapolate to zero is often observed, see for example the measurements of Fe_2O_3 and CrO_2,[20] and indicates that the assumption that $N(E)$ is constant is not a good approximation for these materials. Note that the energy range of barriers sampled depends on the measuring field, measuring temperature, and on the timescale of the experiment. Calculations have been reported for various $N(E)$ distributions and various dependencies of $S(T)$ on temperature[21,22] can be obtained including an $S(T)$ that shows a maximum as a function of temperature. We believe that an appropriate nonuniform distribution of barriers could explain our observed linear $S(T)$ which does not extrapolate to zero.

We now turn to the magnetic relaxation at and below T_0. There is a general correlation between anisotropy strength and T_0 among systems with very weak and with strong anisotropy in this work and in previous work. In Fe systems the magnetic anisotropy tends to be weak (of order 10^5–10^6 ergs/cm^3) and T_0 is in the 0.05 to 2 K range.[2–4] In previous work in rare-earth alloys (Tb-Fe and Dy-Co multilayers and bulk materials) the magnetic anisotropy is strong, of order 10^8 ergs/cm^3, and T_0 values are in the 1–6 K range.[2,3] In previous work within the rare earths, two Tb-based alloys have the highest value of all the rare-earth alloy systems studied, being 6 K for an amorphous TbFe/Cu multilayer[2] and 6 K for an amorphous TbFe alloy.[3] In our work on the elemental Tb, Dy, and Nd multilayers we find a value of 29, 6, and 10 K supporting this general correlation for weak anisotropy (Fe-based) systems and strong anisotropy (rare-earth based) systems and the correlation within the rare earths whereby systems containing Tb tend to have the highest value of T_0. In our $Dy_{50}Co_{50}$/Mo multilayers T_0 is 5.5 K, slightly larger than the highest values previously reported for DyCo alloys. We note that the value of T_0 for the Tb multilayers of this work are unusually large.

The high T_0 in the Tb multilayer allows us to observe the temperature-independent behavior of $S(T)$ over an extended temperature range. We find that the crossover here is sharp with an abrupt change in slope of $S(T)$ at T_0 as has been seen in an amorphous TbFe system at low temperatures.[2,23] Such a sharp crossover is expected in a MQT mechanism if the various dissipations (e.g., interactions of the magnetic system with phonons, magnons) are small as they are expected to be for magnetic systems.[9]

In order to estimate the value of T_0 expected from MQT we use a domain-wall tunneling model[9] with uniaxial anisotropies along the z and y axes of strengths per unit volume $K_\perp$ and $K_\parallel$ (with $K_\perp>K_\parallel$). The pinning of the domain walls in this model is due to the strong magnetic an-

TABLE I. The measured values of $K_{\|}$ and $K_{\perp}$ for bulk Tb and Dy (Ref. 15). The saturation magnetization, measured value of T_0, and estimated value of T_0 from Eq. (2) are also given. The error in the theoretical T_0 is ± 0.2 K and the error in the experimental T_0 is ± 1 K.

	$K_{\|}$ (ergs/cc)	$K_{\perp}$ (ergs/cc)	M_0 (emu/cc)	T_0 (Theory)	T_0 (Experimental)
Tb	2.4×10^6	5.5×10^8	2 700	1.8 K	29 K
Dy	7.5×10^6	5.0×10^8	3 030	2.7 K	6 K

isotropy rather than defects and so is intrinsic. Such a model describes fairly well the magnetic anisotropy of bulk Tb and Dy where the c-axis uniaxial anisotropy ($K_{\perp}$) is strong and the planar anisotropy ($K_{\|}$) is weaker. In this model the crossover temperature is estimated (in the limit of large $K_{\perp}$) to be

$$k_B T_0 = 2\mu_B (K_{\|} K_{\perp})^{1/2} / M_0 , \tag{2}$$

where M_0 is the saturation magnetization per unit volume within a domain. This result is similar to that for magnetic reversal by quantum tunneling of a single domain.[7] Table I gives values for $K_{\|}$, $K_{\perp}$, M_0, and the calculated values of T_0 using Eq. (2) for the Tb and Dy systems. The values of $K_{\|}$ and $K_{\perp}$ are taken from bulk materials[15] and we used these as estimates of the anisotropy constants for our samples. As can be seen, the estimated values of K do not give the observed variation of T_0. The observed maximum value of T_0 for Tb is larger than Dy, while Eq. (2) predicts Tb should have the smaller value. We are unable to identify additional contributions to the anisotropy which might significantly increase the Tb anisotropy over the Dy anisotropy to explain this difference. Thus this simple MQT picture is not able to explain the variation of T_0 in the Tb and Dy multilayers.

We have also observed temperature-independent relaxation at low temperatures in $Dy_{50}Co_{50}$ and in Nd multilayers where the magnetic order is not collinear. An interesting experimental observation is that the value of T_0 was not significantly changed in the Dy multilayers on substituting Co in the form $Dy_{50}Co_{50}$. This substitution modifies two aspects of the system. Firstly it increases the average exchange strength significantly. Secondly since the DyCo layer is amorphous the magnetic anisotropy becomes random in nature on a microscopic scale. Any changes in T_0 due to these two modifications appear to cancel each other for the DyCo system since T_0 is not significantly changed.

A maximum in T_0 was observed as a function of H_m in our Tb multilayer. A similar variation of T_0 with H_m has been reported in a FeTb/Cu multilayer with a maximum value of 6 K.[23] If a MQT interpretation is considered then the decrease of T_0 at high fields observed in this work is expected from a consideration of the energy barriers at high fields. As the field is increased the energy barriers are decreased allowing thermal activation over an energy barrier to occur at lower temperatures. Thus T_0, the temperature below which MQT dominates over thermal activation, is lowered. However we cannot explain the low-field decrease of T_0 (and thus the peak at intermediate fields) in our Tb system in this simple MQT picture.

We now turn to other possible explanations of the temperature-independent magnetic relaxation below T_0. It is clear from the above discussion that a nonuniform distribution of energy barriers is present. A certain distribution of energy barriers, namely a distribution of energy barriers which diverges in a power-law fashion of the form $N(E) \sim 1/E$ over some range of energy, can produce a temperature-independent magnetic relaxation over a temperature range which depends on the energy range.[10,11] It is expected that at low enough temperatures the magnetic relaxation will again become temperature dependent, whereas in MQT the relaxation rate should remain constant as T approaches 0 K. Clearly we cannot eliminate the possibility that the magnetic relaxation becomes temperature dependent at temperatures below those used in this work. It is difficult, however, to see how such a model could lead to a sharp crossover to the temperature-independent regime at T_0 as seen in the Tb system.

A model based on self-organized critical behavior[12] may provide an avenue to explain the above results. An important aspect of such a model is that the components—in our case domain walls making up the system interact with each other. Such interactions should increase with the strength of the exchange interactions between moments. In the rare earths this interaction scales approximately with $S(S+1)$ and so is expected to be larger for Tb than Dy. Thus the temperature above which thermal fluctuations dominate over inter-domain-wall interactions should be higher for Tb systems than Dy systems as is seen in this work. Quantitative comparison is not yet possible since no detailed calculations have been reported on the relaxation of a system of interacting magnetic domains. Any such model should be able to explain the maximum in T_0 as a function of measuring field observed in our work and previous work on Tb systems.

IV. CONCLUSIONS

We have observed a temperature-dependent magnetic viscosity $S(T)$ at high temperatures and at low temperatures (below a T_0 of 29 K for Tb and 6 K for Dy) we see a well-defined crossover to a temperature-independent regime for all of the Tb and Dy rare-earth systems studied. The value of T_0 for the Tb system is the highest value reported thus far for this crossover. The measured values of T_0 for Tb and Dy do not follow the theoretical estimate, Eq. (2), calculated from a simple MQT theory using our estimated magnetic anisotropy values. While the value of T_0 is not explained by the simplest MQT theory, there is a general correlation between anisotropy strength and T_0 with the stronger anisotropy systems (rare-earth-based) having larger values of T_0 and smaller anisotropy systems (Fe-based) having smaller values of T_0. A temperature-independent magnetic relaxation for $T < T_0$ is also present in DyCo/Mo and Nd/Mo multilayers where the magnetic order is nonferromagnetic.

Interesting alternatives to MQT include a distribution of

energy barriers model and an interacting domain-wall model and could explain this temperature-independent magnetic relaxation. In order to explore these alternatives, we are preparing crystalline isolated rare-earth particles in a nonmagnetic matrix with as small a size distribution as possible to eliminate interactions between the relaxing units of the system and to reduce the distribution of energy barriers known to be present in the magnetic systems studied thus far. Possible explanations related to these alternatives can be studied by comparing these systems with the rare-earth layers of this work where the relaxing magnetic units (domain walls) interact with each other and also have a distribution of energy barriers.

ACKNOWLEDGMENTS

We thank E. Chudnovsky for useful discussions on quantum tunneling models and B. Barbara and J. Tejada for discussion of their results. This work was supported by NSF OSR92-55223 and NSF DMR9123831.

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