Slow relaxation of magnetization in MnAs nanomagnets on GaAs(001)

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We demonstrate slow relaxation of the magnetization in MnAs nanoparticles fabricated from epitaxially grown films on GaAs(001). In disks having a diameter as small as 100 nm and a thickness of 50 nm, the decay of the magnetization at 27 °C, which is merely ~10 °C below the Curie temperature T_C, is less than 1% over a period of one day. The large uniaxial magnetocrystalline anisotropy and the abrupt loss of the ferromagnetism at T_C of MnAs are responsible for the slow relaxation. © *2006 American Institute of Physics*.
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In the course of miniaturizing a unit cell of magnetic storages, the associated magnetic energies eventually become smaller than the thermal energy k_BT. The magnetization of a particle having a uniaxial magnetic anisotropy can thus flip to the opposite direction by thermally surmounting an energy barrier. This instability imposes a limit to the data storage density, which has increased steadily since the 1950s at a rate of tenfold in 6 years.[1,2] There is a number of proposals to extend the limit. One intriguing example is the use of the exchange biasing effect to suppress the superparamagnetism.[3] Even within a conventional approach, one can, in principle, avoid the thermal instability by choosing materials having a large anisotropy constant K, as thermal agitation is a function of a quantity KV/k_BT, where V is the particle volume.[2] However, K cannot be too large in practice as the external magnetic field required to record magnetic data scales in proportion to K. Thermally assisted magnetic recording, for instance, is a compromising technique, in which the recording is carried out at an elevated temperature to reduce the coercivity.[4]

In this letter, we demonstrate a markedly slow relaxation of the magnetization in MnAs nanoparticles resulting from the large K value ($=7\times10^5$ J/m^3 at room temperature[5]) of the material. (Note for comparison that $K=4.8\times10^4$ J/m^3 in bcc Fe.) MnAs is attractive also from the viewpoint of the thermally assisted magnetic recording as the Curie temperature T_C of bulk MnAs is only about 40 °C. In spite of the long-term stability of the magnetization at room temperature, the coercivity is diminished by increasing the temperature by a mere several tens of a degree.

We investigate the magnetization in MnAs disks fabricated from two 50-nm-thick films. The MnAs layers were grown on GaAs(001) substrates using molecular beam epitaxy.[6] The growth conditions were optimized to have the surface of the epitaxial MnAs layers being oriented as $(1\bar{1}00)$.[7] The [0001] and $[11\bar{2}0]$ directions of MnAs were aligned along the $[1\bar{1}0]$ and [110] directions of GaAs, respectively. The MnAs/GaAs heterostructures were processed to the arrays of MnAs disks using microfabrication technologies. A NiCr etch mask was prepared on the MnAs surface using electron beam lithography and the lift-off technique. The MnAs layers were transformed into disks depicted by the mask using Ar ion milling at an acceleration voltage of 150 V. We examine two samples, of which the scanning electron micrographs are shown in Fig. 1. The magnetization of the samples was measured using a superconducting-quantum-interference-device magnetometer (Quantum Design MPMS XL). The metal mask was left on top of the MnAs disks during the measurements. We have confirmed that the deposited NiCr is paramagnetic, and so it does not affect the relaxation of the magnetization in the MnAs disks.

In Fig. 2, we show the magnetization curve of the MnAs disks shown in Fig. 1(a) at a temperature $T=20$ °C. The diameter of the disks is $d=100$ nm. The external magnetic field H is applied along the magnetic easy axis, which is along the $[11\bar{2}0]$ direction of MnAs. The magnetization M is normalized by the saturation magnetization M_s. For comparison, the magnetization curves of the MnAs film from which the sample was fabricated are also plotted. Here, H is applied along the MnAs$[11\bar{2}0]$ direction (easy axis) for the dotted line and along the MnAs[0001] direction (hard axis) for the dashed line. Notice that the uniaxial magnetocrystalline anisotropy in MnAs films is remarkably large. As we show later, the large anisotropy leads to unique magnetic properties of MnAs disks. The coercivity of the disks (0.73 kOe) is similar to that of the film. In the disks, the remanence M_r is only 20% smaller than M_s. The similarity of the magnetization curve of the disks to that of the film suggests that the thermal effects on the magnetic moments are considerably small in the MnAs disks.

Before we analyze the magnetic properties of the disks further, let us describe the behavior of the magnetic domains

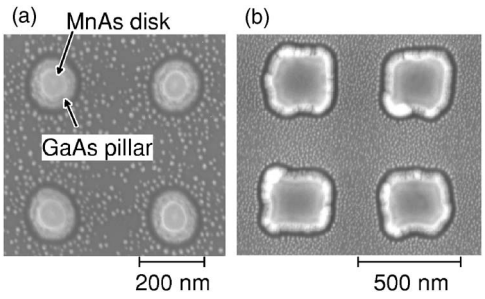

FIG. 1. Scanning electron micrographs of MnAs disks fabricated from 50-nm-thick films grown on GaAs(001). The size of the disks is (a) 100 and (b) 200 nm. The two samples were processed from separate films. The MnAs disks sit on top of GaAs pillars as the Ar ion milling proceeded about 100 nm deep into the substrates. The square arrays of the disks were defined along the cleavage directions of GaAs.

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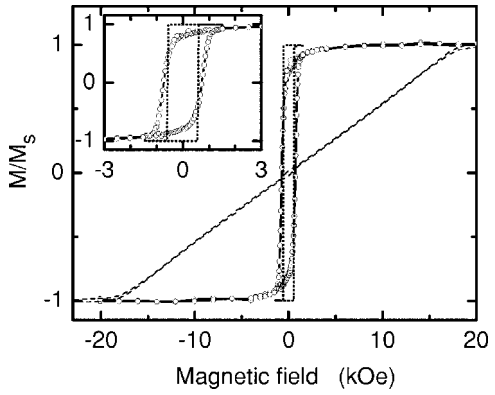

FIG. 2. Magnetization curves of MnAs film and disks fabricated from the film. The magnetization M is normalized to the saturation magnetization M_s. The in-plane external magnetic field is applied for the film along the uniaxial easy axis, MnAs[$11\bar{2}0$] (dotted line), or the hard axis, MnAs[0001] (dashed line). For the disks having the size of 100 nm, Fig. 1(a), the magnetic field is applied along the easy axis (solid line with circles). The hysteresis loops are shown with an expanded scale in the inset.

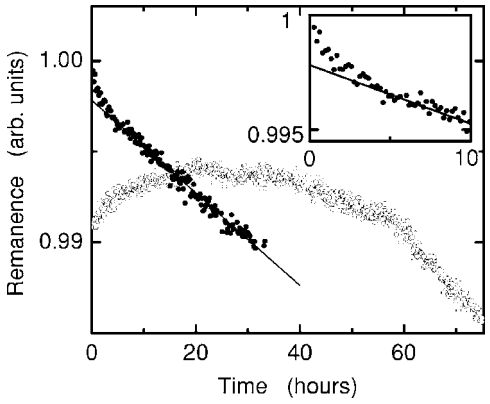

FIG. 3. Time dependence of the remanence, normalized by a common value, in disks having a diameter of 100 nm, Fig. 1(a). The disks were magnetized prior to the measurements in a magnetic field of 20 kOe. The temperature is 27 and 20 °C for the filled and open circles, respectively. The sample was at a lower (higher) temperature than the measurement temperature for the filled (open) circles immediately before the measurements. The solid line is a fit assuming an exponential decay, Eq. (1). The inset is an expanded plot to show a rapidly decaying component.

in submicrometer disks. The magnetic domain structure undergoes a fundamental change when the dimensions of ferromagnetic elements are reduced to nanometer scales. That is, a transition between a vortex state and a single-domain state occurs.[8] In the disks larger than a critical size, the magnetic structure is a vortex (closure-flux-type multidomain) state,[9] unless the magnetic properties of the ferromagnetic material are highly anisotropic. When the disk size is below the critical value, however, the domain-wall energy exceeds the magnetostatic energy arising from stray fields. The reversal in the energy relationship originates from the nonzero width of the domain walls. A single magnetic domain thus occupies the entire disk in the latter regime even after demagnetization. We have established using magnetic force microscopy that the critical size to reach the single domain regime is about 100 nm for the present MnAs layer thickness.[10]

We now turn our attention back to the relaxation of the magnetization in the disks. In the sample with $d=100$ nm, most of the disks are single domain particles.[10] The relaxation of the magnetization in these so-called nanomagnets arises from the probabilistic process of the thermally induced reversal of their magnetic moments. The decay of the magnetization with time t is hence given as

$$M_r(t) = M_r(0)\exp(-2t/\tau), \tag{1}$$

where τ^{-1} is the flipping rate of the magnetic moments. We show in Fig. 3 the time dependence of the magnetization at $H=0$ after the MnAs disks were magnetized at $H=20$ kOe. One finds that the experimental results are not as simple as predicted by Eq. (1).

The sample temperature was 27 and 20 °C for the filled and open circles, respectively. However, the qualitatively different time dependence in the two measurements is a consequence of the contrasting temperature at which the sample dwelled prior to the measurements. The sample was warmed to the measurement temperature for the filled circles, whereas the sample temperature was 32 °C before the measurement for the open circles. The magnetization initially increased in the measurement run shown by the open circles. In contrast, a rapid decay (the deviation from the extrapolation of the long-time behavior indicated by the solid line in the inset of Fig. 3) was observed in the measurement run shown by the filled circles. Through measurements under various thermal histories, we have established that the increase and the rapid decrease in the magnetization take place when the samples are cooled and warmed to the measurement temperature, respectively. We, therefore, attribute these rapidly quenching contributions to the temperature hysteresis of the first-order phase transition between the ferromagnetic α phase and the nonmagnetic β phase of MnAs at T_C ($\approx$40 °C).

In a wide temperature range around T_C, which includes room temperature, both of the disks consisting of α-MnAs and of β-MnAs exist when the material is grown on GaAs.[10] This type of coexistence is a manifestation that a first-order phase transition does not occur immediately when the temperature is varied from a value favoring one phase to another value favoring the other phase. An abrupt phase transition takes place in each disk probabilistically by surmounting the potential barrier separating the phases. The thermal hysteresis thus gives rise to a relaxation behavior. We note, however, that some characteristics of the rapid decay are not clearly understood at present by the interpretation based on the thermal hysteresis. (i) The increase of the magnetization implies that the newly converted α-MnAs disks are, at least predominantly, magnetized in the direction of the magnetic moments of the surrounding disks. Dipole-dipole interaction might be responsible for the magnetic ordering. (ii) The initial increase ($\beta \rightarrow \alpha$ transition) takes place on a time scale which is much longer than that when the counterpart ($\alpha \rightarrow \beta$ transition) leads to a decay of the magnetization. The asymmetry may be intimately related to (i), i.e., the $\alpha \rightarrow \beta$ transition always results in a decrease of the magnetization, whereas the $\beta \rightarrow \alpha$ transition does not have to increase the magnetization.

Once the fast-relaxation contributions diminish, the magnetization decays at a comparable rate for both of the measurement runs in Fig. 3. We attribute this slow decay to the thermal switching of the magnetic moments. If we assume a relaxation given by Eq. (1) (the solid line in Fig. 3), the relaxation time is estimated to be $\tau \sim 300$ days. The switching events of the magnetization are hence indicated to

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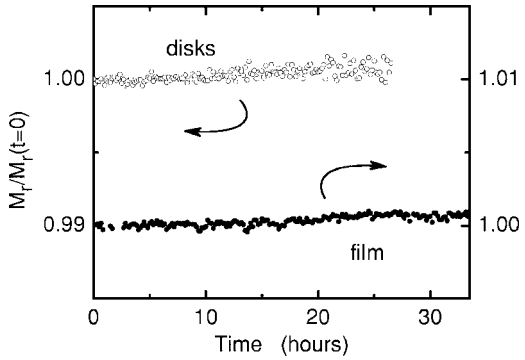

FIG. 4. Time dependence of the remanence M_r normalized to the value at time $t=0$ at 20 °C. The open and filled circles correspond to disks having a size of 200 nm, Fig. 1(b), and the MnAs film from which the disks were fabricated, respectively. The samples were magnetized at 5 kOe prior to the measurements.

be scarce in MnAs disks owing to the gigantic uniaxial magnetocrystalline anisotropy.

In general, magnetic moments thermally fluctuate around the easy axis. The extremely slow relaxation suggests that the deviation angles are restricted to be fairly small in MnAs disks and the thermal effects provide no appreciable contribution for lowering M_r below M_s. The difference between M_r and M_s in Fig. 2 is hence ascribed to a distortion of atomic magnetic moments by magnetic interactions, i.e., a "spin texture" that develops near the edges of the disks.[11]

Figure 4 shows the time dependence of the magnetization in disks having $d=200$ nm, see Fig. 1(b). For this disk size, the two phases of MnAs coexist within a single disk, presumably in the form of a core-shell-type phase segregation.[10] With respect to the magnetic properties, on the one hand, the magnetic energy is estimated to be several times larger in this sample than in the sample in Fig. 3, thereby stabilizing the magnetization. On the other hand, as the disks are in the multidomain regime, the magnetization can relax not only by flipping the total magnetic moment but also by creating a domain wall, i.e., reversing a part of the magnetization. The disks are expected to contain no more than two domains, as the large magnetocrystalline anisotropy in MnAs$(1\bar{1}00)$ films disfavors a tilt of the magnetization away from the easy axis caused by the shape anisotropy.[10] The relaxation would not be enhanced by a domain-wall movement. The experimental result demonstrates that the relaxation is, at least, comparably slow in the multidomain disks.

To overcome the recording magnetic field limit which is encountered when a high-K material is used, thermally assisted magnetic recording has been proposed.[4] MnAs-based devices are prospective for such applications as, despite the thermal stability of the magnetization at room temperature, T_C of the material can be exceeded by increasing the temperature by several tens of a degree. We also emphasize a unique magnetic property of MnAs, i.e., the loss of the ferromagnetism at T_C is discontinuous.[12] The magnetic moment remains unusually large even for temperatures slightly below T_C, and the magnetic anisotropy can be substantial to prevent the thermal randomization, as we demonstrated in the present work. Although T_C of bulk MnAs may be too low for some applications, we point out that the phase transition temperature in epitaxial films grown on substrates can be manipulated by strain control. The transition temperature in disks can be raised by enhancing the tensile strain due to the thermal expansion mismatch between MnAs and the substrate material. In addition, the thermal hysteresis could be suppressed by optimizing the growth conditions.[13]

In conclusion, we have evaluated the relaxation time of the magnetization in 100- and 200-nm-large MnAs disks on GaAs(001). The uniaxial magnetocrystalline anisotropy in MnAs is found to be large enough to stabilize the magnetization even at room temperature. The thermal stability is maintained even though the temperature is merely 15–20 °C below the Curie temperature of MnAs.

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