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Suppression of the increase of high-temperature coercivity in MnBi thin films by Al interlayers

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By tailoring the microcrystalline structure of MnBi films, using Al interlayers, a reduction of the high-temperature coercivity by a factor of 3 is achieved. The separation of Bi/Mn bilayers by Al interlayers acts as a diffusion barrier perpendicular to the surface. After annealing, the MnBi layers contain single-domain particles surrounded by an Al matrix exhibiting no significant increase of the coercive field with increasing temperature.
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The large negative Kerr rotation of nearly $\Theta_K = -1.2°$ in the UV spectral region and the large perpendicular magnetic anisotropy make MnBi a promising material for high-density, short-wavelength magneto-optic recording.[1] A laser beam is commonly used for reading and writing. As a consequence, the smallest diffraction-limited spot-size diameter is 0.56 λ/NA. The spot size can be reduced not only by decreasing the wavelength λ but also by increasing the numerical aperture NA. The recently developed blue semiconductor lasers with $\lambda = 417$ nm reduce the theoretical spot-size minimum to nearly 240 nm. Therefore, in order to get a low-noise readout signal, the diameter of the crystallites has to be minimized to sizes smaller than 25 nm. A MnBi grain size of more than ≈ 1 μm in diameter gives rise to a small signal-to-noise ratio of the readout signal.[1] The grain size reduces by adding a small content of Al, Sb or Pt to the MnBi films.[2,3] For example, MnBi films with Al interlayers exhibit grain sizes smaller than about 100 nm in diameter.[2]

An unusual increase of the coercive field, H_c, reaching 2.0 T at 550 K is detrimental to the thermomagnetic writing process.[4] Several years ago, a hybrid domain-wall-pinning model was proposed to explain the temperature dependence of the coercive field.[5] The main feature of this model is the thermal activation of domain walls pinned by nonmagnetic defects or impurities during the magnetization-reversal process. In the case of single-domain particles, a temperature-dependent pinning mechanism of domain walls is not important. By comparing the magnetic energy of a single-domain MnBi sphere with that of a sphere showing internal flux closure, the upper limit in size for a single-domain state is estimated to be nearly 100 nm. For calculating this critical diameter a Bloch-wall energy of $S_B = 0.016$ J/m^2, a saturation magnetization of $M_S = 660$ kA/m, and a uniaxial anisotropy constant $K_u = 1160$ kJ/m^3 were used.[5] In this letter, we show that adding Al interlayers to MnBi in a suitable way will reduce MnBi grain size below 100 nm and, therefore, effectively suppress the domain-wall pinning mechanism. This leads to a vanishing of the strong increase of H_c between room temperature and 550 K.

A sequence of three Bi (18 nm)/Mn (12 nm) bilayers has been deposited on quartz substrates in a vacuum of $\sim 10^{-6}$ mbar. The deposition rates of the Bi and Mn layers were fixed to 0.4 and 2.0 Å/s, respectively.[6] The Bi/Mn bilayers are separated from each other by 2.0-nm-thick Al interlayers and covered by a 2.0-nm-thick Al capping layer. In order to get c-axis oriented MnBi films the multilayers were annealed for 30 min at 653 K and for 30 min at 593 K. During annealing the vacuum stayed in the range from 10^{-5} mbar. After annealing, the structural and magnetic properties of the MnBi films were characterized by x-ray diffraction analysis (XRD), Rutherford backscattering spectrometry (RBS), transmission electron microscopy (TEM), polar Kerr hysteresis-loop measurements at a photon energy of 2.0 eV and polar Kerr spectroscopy in a photon-energy range of 0.8–5.2 eV. The temperature dependence of the coercive field, H_c, was determined in a quartz tube in a vacuum of 10^{-5} mbar. The samples were mounted on a heatable copper rod which allows for a temperature adjustment up to 730 K with an uncertainty of ± 5 K. Details of the preparation procedure and the structural properties of MnBi and MnBiAl films were published elsewhere.[6]

The temperature dependence of the coercive field, H_c, and the saturation magnetization, M_s, as obtained from polar Kerr hysteresis loops, are plotted in Fig. 1. In contrast to pure MnBi films of similar thickness without Al interlayers[7] a larger H_c of 0.48 T at 293 K and a broad maximum of only 0.6 T in the temperature range near 450 K is observed. For comparision, a value of H_c=1.9 T is reported for pure MnBi at 550 K.[5] For temperatures higher than 500 K, H_c and M_s drop rapidly up to 660 K, when the phase transition to the nonferromagnetic high-temperature phase of MnBi occurs. Following the magnetization curve the small kink at 500 K is explained by an additional MnBi-formation process during the high-temperature measurement.

Due to the additional MnBi formation the maximum polar Kerr rotation increases from $-0.8°$ to $-1.15°$ after the HT measurement as demonstrated in Fig. 2. The enhance-

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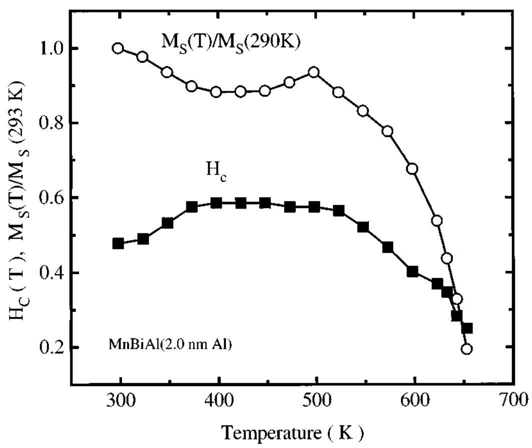

FIG. 1. Temperature dependence of the coercive field, H_c, and saturation magnetization, M_s, of a MnBiAl (2.0 nm) film.

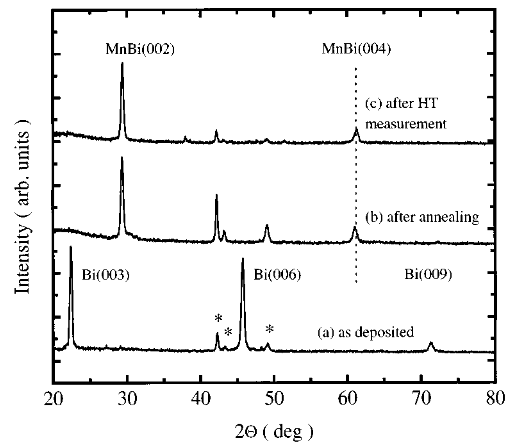

FIG. 3. X-ray pattern of (a) an as-deposited Bi/Mn/Al multilayer, (b) a Mn/Bi/Al (2.0 nm) film after annealing, and (c) after HT measurement.

ment of the polar Kerr rotation after the HT measurements is based on a small increase of the saturation magnetization M_S. Nevertheless, the coercive field of 0.5 T and, as a consequence, the microcrystalline structure of MnBi particles do not change.[6,7] The rectangular-shaped hysteresis loops exhibit a squareness of nearly one.

X-ray diffraction analysis has been performed in order to exclude irreversible changes during the HT cycles like phase separation into a Bi and a MnBi phase or partial oxidation of the MnBi film. The x-ray pattern of the Bi/Mn/Al (2.0 nm) multilayers in Fig. 3(a) shows only Bi(00ℓ) reflexes indicating full c-axis texture of the Bi layers after deposition. Peaks due to the brass substrate holder, are labeled with an asterisk. After annealing, the x-ray pattern in Fig. 3(b) exhibits only MnBi(00ℓ) reflexes. The x-ray pattern in Fig. 3(c) of the same MnBi film performed after HT measurement shows no additional peaks besides the MnBi(00ℓ) reflexes indicating that no irreversible processes like phase separation into pure Bi and MnBi or partial oxidization occur. In case of only 0.2-nm-thick Al interlayers a phase separation into Bi and MnBi was observed after the HT cycles.

In a previous article a reduced diffusion of the Bi/Mn bilayers across Al interlayers during annealing has been discussed.[6] The diffusion profile across the Bi/Mn/Al stacks was studied by RBS measurements using 1.4 MeV He^+ ions. Details of the experimental setup are given in Ref. 6. Before annealing, a clear separation of the individual Bi and Mn layers was observed as a double-peak structure. Without an Al interlayer the double-peak structure of the two Bi and Mn layers disappeared completely after annealing, whereas for a 2.0-nm-thick Al interlayer the double-peak structure for Bi and Mn remained.[7] This means that, due to the 2.0-nm-thick Al interlayers, two individual MnBi layers have been formed. The Al interlayer, which is probably partially oxidized, acts as diffusion barrier. In contrast, for Al interlayers smaller than ≈ 1.4 nm no separation into two individual MnBi layers was observed.

From RBS-, TEM-, and polar Kerr hysteresis-loop measurements a microcrystalline model of the multilayered MnBi films after annealing is developed as shown in Fig. 4. During annealing the growth of MnBi crystallites across the Al interlayers perpendicular to film surface is reduced, whereas the growth parallel to each Bi/Mn bilayer is not restricted. A small amount of the Al interlayers diffuses into the MnBi grain boundaries. Caused by the Al interlayers, three individual MnBi layers arise consisting of lens-shaped MnBi grains. The diameter of the MnBi grains is estimated by TEM measurements to be smaller than 100 nm. As mentioned above, this grain size is the upper limit for a single-domain state. The small MnBi crystallite size, i.e. the single-domain state of the grains, accounts for the large coercive field of 0.48 T in comparison to pure MnBi films at room temperature.[7] The single-domain state of each MnBi grain suppresses, on the other hand, the increase of the coercivity at high temperatures, which had been explained by a

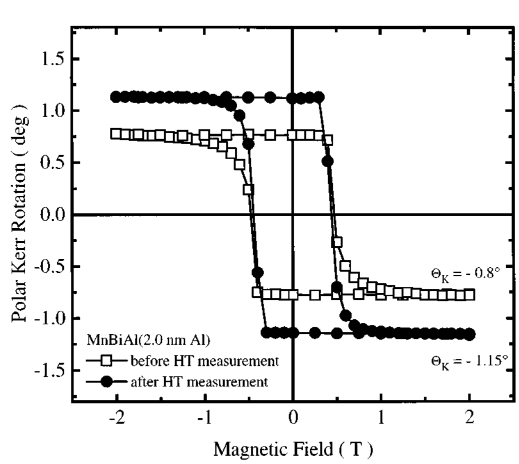

FIG. 2. Polar Kerr hysteresis loops before and after a high-temperature measurement.

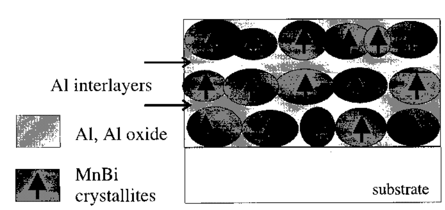

FIG. 4. Schematic microcrystalline model of a Bi/Mn/Al multilayer after annealing.

temperature-dependent domain-wall-pinning mechanism.[5] In contrast to pure MnBi films, consisting of multi-domain grains, this temperature-dependent domain-wall-pinning mechanism is not present in multilayered MnBi films and, therefore, no strong increase of the coercive field with increasing temperature is observed.

In conclusion, the microcrystalline structure of MnBi films, i.e. the magnetic domain configuration of the MnBi crystallites, seems to be the origin of the unusually large coercivity of pure MnBi films at high temperatures. Reducing the MnBi grain size by separating Bi/Mn bilayers by sufficiently thick Al interlayers leads to a single-domain state of nearly each MnBi particle. Due to the single-domain state, pinning mechanisms of domain walls are not active and, therefore, the coercive field does not exhibit a strong temperature dependence.

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