

# Size dependence of the exchange bias field in NiO/Ni nanostructures

M. Fraune, U. Rüdiger,<sup>a)</sup> and G. Güntherodt  
*II. Physikalisches Institut, RWTH Aachen, 52056 Aachen, Germany*

S. Cardoso and P. Freitas  
*INESC, Rua Alves Redol 9, 1000-029 Lisboa, Portugal*

(Received 14 August 2000; accepted for publication 16 October 2000)

NiO/Ni wires have been investigated as a function of their width in order to investigate the size dependence of exchange bias. The samples have been prepared by e-beam lithography and ion milling of ion beam sputtered thin films. For NiO/Ni wires narrower than 3  $\mu\text{m}$ , the exchange bias field significantly depends on the wire width. A NiO/Ni film shows an exchange bias field of  $-78$  Oe whereas the exchange bias field of wires narrower than 200 nm is reduced to approximately  $-40$  Oe. The coercive field of the NiO/Ni film is 28 Oe and increases to 210 Oe for the narrowest wires. The decrease of the exchange bias field for the narrowest wires is consistent with a recent microscopic model of exchange bias where the appearance of a unidirectional anisotropy in ferromagnet/antiferromagnet bilayers has been attributed to the presence of antiferromagnetic domains in the bulk of the antiferromagnet. A possible onset of a transition from a multidomain to a single-domain state of the antiferromagnet as a function of the NiO/Ni wire width seems to be the origin for the observed decrease of the exchange bias field for narrow wires.

The exchange bias (EB) effect occurs due to the exchange coupling at antiferromagnet(AFM)/ferromagnet(FM) interfaces leading to a shift of the magnetic hysteresis loop along the field axis.<sup>1</sup> This shift of the hysteresis loop can be established either by cooling the AFM/FM bilayers in a magnetic saturation field below the Néel temperature  $T_N$  of the AFM or by depositing the bilayers in an external magnetic field.<sup>2</sup> The exchange bias effect has been used over several decades and more recently for pinning the magnetization of one of the two electrodes in magnetoresistive devices (giant magnetoresistance multilayers<sup>3</sup> and tunnel junctions<sup>4,5</sup>). Although the exchange bias effect has already been intensively exploited in micron- and submicron-sized magnetoelectronic devices, its microscopic origin is not yet fully understood.

A recent experiment on Co/CoO bilayers in conjunction with a Monte Carlo simulation study has shown that the dilution of the antiferromagnet CoO with nonmagnetic impurities (e.g.,  $\text{Co}_{1-x}\text{Mg}_x\text{O}$ ) or defects (e.g.,  $\text{Co}_{1-y}\text{O}$ ) in its volume part leads to the formation of antiferromagnetic volume domains.<sup>6</sup> The formation of antiferromagnetic domain walls leads to a small surplus magnetization at the AFM/FM interface which couples to the FM and results in a unidirectional anisotropy. Hence, the antiferromagnetic domains are the microscopic origin of exchange bias. This result is complementary to a previous approach attributing exchange bias to the formation of antiferromagnetic domains with domain walls (DW) perpendicular to the AFM/FM interface in the presence of only interface roughness.<sup>7</sup> From these models one has to conclude that the exchange bias of AFM/FM bilayers vanishes when the AFM becomes single domain. From a systematic investigation of a possible finite size effect of exchange bias the microscopic role of the DW forma-

tion can be elucidated as well as the lower limit of the extension of EB-based devices can be estimated. Relatively large AFM domain sizes have been visualized in NiO films<sup>8,9</sup> and NiO single crystals<sup>10</sup> by x-ray magnetic linear dichroism spectromicroscopy.

Although much experimental work has been done on the topic of exchange bias, finite size effects on a 100 nm lateral scale have not yet been investigated systematically.<sup>11</sup> In this letter, NiO(40 nm)/Ni(10 nm) bilayers are used as a test system for the investigation of exchange bias as a function of laterally reduced dimensions.

The antiferromagnet NiO crystallizes in the rock-salt structure with a lattice constant of  $a_0 = 4.17 \text{ \AA}$ .<sup>12</sup> In NiO the spins are ferromagnetically coupled within the (111) planes whereas the spins of alternating (111) planes are antiferromagnetically coupled.<sup>13</sup> The antiferromagnetic ordering temperature is  $T_N = 523 \text{ K}$ .<sup>12</sup> The exchange constant for the nearest neighbor (NN) interaction is  $J_1 = -4.7 \times 10^{-22} \text{ J}$ <sup>12</sup> and for the next nearest neighbor interaction  $J_2 = -2.8 \times 10^{-21} \text{ J}$ ,<sup>13</sup> which leads to exchange stiffness constants of  $A_1 = 1.6 \times 10^{-12} \text{ J/m}$  and  $A_2 = 6.7 \times 10^{-12} \text{ J/m}$ . With the magnetic anisotropies of  $K_{\perp} = 3.3 \times 10^5 \text{ J/m}^3$ <sup>13</sup> perpendicular to the (111) plane and  $K_{\parallel} = 33 \text{ J/m}^3$ <sup>14</sup> parallel to the (111) plane the antiferromagnetic domain wall widths can be estimated to  $D_{\perp} = \pi \sqrt{A_2/K_{\perp}} = 14.2 \text{ nm}$  perpendicular and  $D_{\parallel} = \pi \sqrt{A_2/K_{\parallel}} = 1.4 \mu\text{m}$  parallel to the (111) plane of NiO, respectively. Calculating the domain wall widths, the smaller NN exchange stiffness constant  $A_1$  has been neglected. The large AFM domain wall width and, therefore, domain size parallel to the (111) plane is in agreement with a recent photoemission electron microscopy experiment on NiO single crystals, where domain sizes of 1–10  $\mu\text{m}$  have been observed.<sup>10</sup> In NiO antiferromagnetic domains have to be at least significantly larger than the AFM domain wall width. The relatively small anisotropy constants of NiO and, there-

<sup>a)</sup>Author to whom all correspondence should be addressed; electronic mail: ruediger@physik.rwth-aachen.de

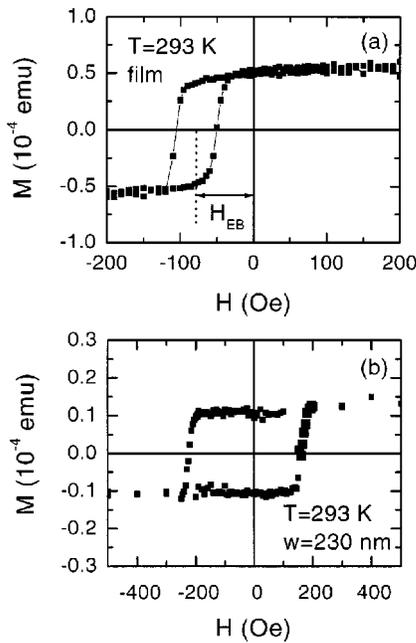


FIG. 1. Magnetic hysteresis loop measurements at  $T=293$  K of (a) an unpatterned NiO(40 nm)/Ni(10 nm) film and (b) an array of 230 nm wide wires.

fore, large antiferromagnetic domain sizes (large domain wall widths) in comparison with other antiferromagnets, make NiO an ideal candidate for an experimental investigation of the effect of laterally reduced dimensions on the exchange bias field of NiO/Ni bilayers.

Polycrystalline NiO(40 nm)/Ni(10 nm) bilayers were deposited on SiO<sub>x</sub>/Si(001) substrates applying an ion beam sputtering technique.<sup>15</sup> The NiO layers were prepared by ion beam sputtering from a 99.8% pure Ni target in the presence of a mixed Ar–O<sub>2</sub> beam coming from an assist gun. For the deposition gun an acceleration voltage of +1450 V (29 mA Xe beam, 1 sccm Xe) and an extraction voltage of –300 V were used. The Ar–O<sub>2</sub> beam was generated by accelerating the plasma inside the assist gun with +200 V (18 mA beam, 10 sccm Ar–O<sub>2</sub>) and using an extraction voltage of –100 V. The pressure during the process was  $1.6 \times 10^{-4}$  mbar. A filamentless neutralizer (40 mA, 3 sccm Ar–O<sub>2</sub>) beside the assist ion source avoids surface charging during the deposition of NiO. During the NiO deposition the substrate was heated to approximately 80 °C and rotated at 15 rpm. The Ni layers were prepared by ion beam sputtering with a pure Ar beam.

An x-ray diffraction analysis shows the polycrystalline character of the NiO(40 nm)/Ni(10 nm) bilayers. Atomic force microscopy of the NiO/Ni film gives an average grain size of the NiO layer of approximately 50 nm and a root-mean-square roughness of 0.24 nm. Comparing the NiO grain size with the antiferromagnetic domain size assumed in first approximation to be proportional to the domain wall width one can conclude that individual NiO crystallites are probably single domain. The antiferromagnetic domain walls are—due to reduced magnetic exchange—located near grain boundaries, minimizing the total energy of the system.

The exchange bias field  $H_{EB}$  was set by heating the NiO/Ni bilayers to  $T=553$  K, which is sufficiently larger than the Néel temperature of  $T_N=523$  K, and cooling to 293 K in a magnetic field of  $H=10$  kOe. Figure 1(a) shows a

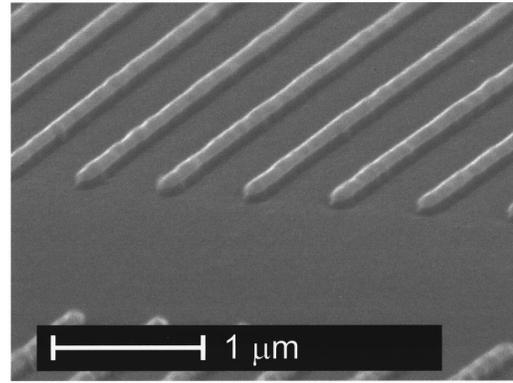


FIG. 2. Scanning electron microscopy (SEM) image of an array of 190 nm wide NiO(40 nm)/Ni(10 nm) wires with a periodicity of 600 nm.

magnetic hysteresis loop measurement of a NiO(40 nm)/Ni(10 nm) film at 293 K using a superconducting quantum interference device (SQUID) magnetometer. At  $T=293$  K the exchange bias field has been determined to  $H_{EB} = -78$  Oe and the coercive field to  $H_c = 28$  Oe. At  $T=5$  K the exchange bias field and the coercive field increase to  $H_{EB} = -258$  Oe and  $H_c = 378$  Oe, respectively. Before cooling to 5 K the Ni layer was always saturated in the same magnetic field direction as during setting the exchange bias in order to avoid misinterpretations resulting from the appearance of memory effects.<sup>16</sup>

From the NiO/Ni bilayers, wires have been nanofabricated by electron beam lithography in conjunction with ion milling using Ar<sup>+</sup> ions. In Fig. 2 a scanning electron microscopy image of an array of 190 nm wide NiO/Ni wires with a periodicity of 600 nm is shown, where the NiO top layer appears in a brighter contrast than the underlying Ni and Si layer. Due to the small saturation magnetization of Ni (493 emu/cm<sup>3</sup>) large area (5 mm×6 mm) NiO/Ni wire arrays have been fabricated to achieve a sufficient signal-to-noise ratio for their magnetization reversal in SQUID measurements. The separation of the wires is always more than twice the wire width  $w$  in order to avoid magnetic dipole interactions,<sup>17</sup> which could have a significant influence on the magnetization reversal process.

Figure 1(b) shows a magnetization reversal loop at  $T=293$  K of an array of 230 nm wide wires with a wire length of  $L=100$  μm and a periodicity of 800 nm. The exchange bias field has been determined to  $H_{EB}=31$  Oe and the coercive field to  $H_c=191$  Oe. In comparison with the unpatterned NiO(40 nm)/Ni(10 nm) film the exchange bias field of this array is reduced to less than half the initial value. The increase of the coercive field by a factor of seven is obvious and is a result of the large shape anisotropy of the wires with an aspect ratio of  $L/w=435$ . Figure 3 summarizes the width dependence of (a) the exchange bias field  $H_{EB}$  and (b) the coercive field  $H_c$  at  $T=293$  K. For wire widths narrower than 1000 nm the exchange bias field decreases to values smaller than  $H_{EB}=-40$  Oe and the coercive field continuously increases up to  $H_c=225$  Oe for the narrowest wire width of 110 nm due to the increasing shape anisotropy. The dotted line in Fig. 3(b) is a fit of the  $L/w$  dependence of the coercive field  $H_c$  ( $H_c=aL/w$ ) showing that the increase of the coercivity is related to an  $L/w$  dependence of the shape anisotropy of the wires.

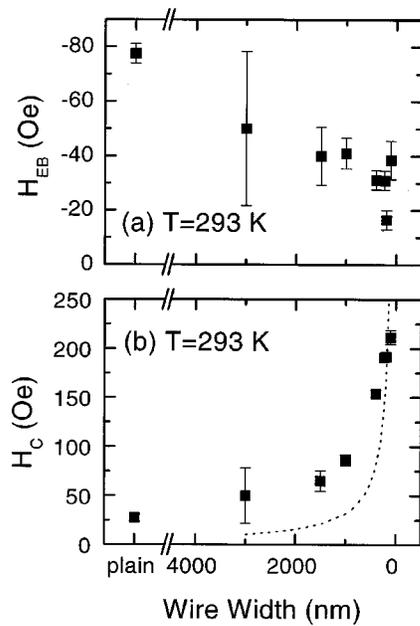


FIG. 3. Exchange bias field (a) and coercive field (b) at  $T=293$  K of NiO(40 nm)/Ni(10 nm) wires as a function of their width. The dotted line is a fit of the coercive field  $H_c$  as a function of the wires aspect ratio ( $H_c = aL/w$ ).

The same determination of the exchange bias field and the coercive field of these wires has been carried out at  $T=5$  K (see Fig. 4). At this temperature the exchange bias field does not show a significant dependence on the wire width as shown in Fig. 4(a). Especially for the narrowest wire widths, no clear reduction of the exchange bias field has been observed. For the smallest wire width ( $w=110$  nm) the exchange bias field of  $H_{EB} = -305$  Oe is even a little bit larger than for the unpatterned NiO/Ni film with  $H_{EB} = 258$  Oe. From this observation at low temperature one can conclude that structural changes at the AFM/FM interface or in the bulk of the AFM due to the ion milling process<sup>18</sup> are

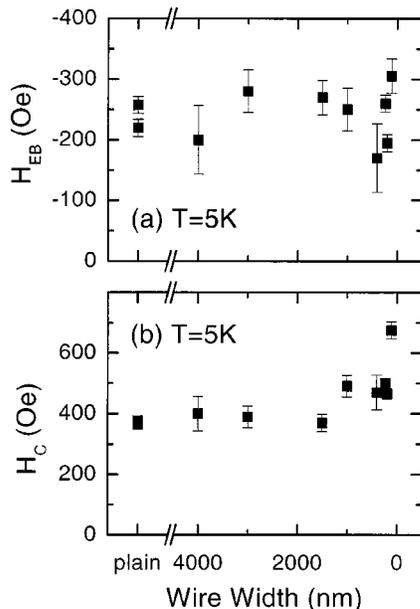


FIG. 4. Exchange bias field (a) and coercive field (b) at  $T=5$  K of NiO(40 nm)/Ni(10 nm) wires as a function of their width.

not responsible for the reduced exchange bias observed for small wire widths at  $T=293$  K. The coercive field as function of the wire width [see Fig. 4(b)] exhibits a similar dependence on the aspect ratio  $L/w$  as observed at  $T=293$  K. As a consequence of the high coercivity of the unpatterned bilayers at  $T=5$  K, the contribution of the shape anisotropy becomes visible only for small wire widths.

The origin of the overall decrease of exchange bias for fixed wire width with increasing temperature is due to the reduction of the surplus magnetization at the AFM/FM interface due to thermal fluctuations as well as the thermally activated depinning of antiferromagnetic domain walls. The wire widths of the narrowest NiO/Ni wires investigated are, within a factor of two, comparable with the NiO crystallite size, and therefore, these wires should tend towards a single-domain state. Thermal activation of pinned antiferromagnetic domain walls in these wires with only a few antiferromagnetic domain walls per wire width will have a stronger influence on the magnitude of the exchange bias field than in unpatterned films. This explains the observed reduction of the exchange bias field at  $T=293$  K and the almost constant exchange bias field at  $T=5$  K. For a final proof, NiO/Ni wires of widths significantly smaller than the NiO crystallite size have to be prepared but this reaches the limits of present microfabrication techniques.

In summary, a significant reduction of the exchange bias field of NiO/Ni nanowires has been observed at  $T=293$  K, compared to unpatterned films. This size effect is consistent with a recently proposed model for exchange bias<sup>6</sup> in which nonmagnetic defects in the volume part of the AFM lead to the formation and stabilization of AFM domains. These domains are considered to be responsible for the exchange bias. The onset of a transition from a multidomain to a single-domain state in NiO seems to be the origin of the observed decrease of the exchange bias field in the NiO/Ni nanowires.

<sup>1</sup>W. H. Meiklejohn and C. P. Bean, Phys. Rev. **105**, 904 (1957).

<sup>2</sup>J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999).

<sup>3</sup>B. Dieny, V. S. Speriosu, S. S. P. Parkin, B. A. Gurney, D. R. Wilhoit, and D. Mauri, Phys. Rev. B **43**, 1297 (1991).

<sup>4</sup>W. J. Gallagher, S. S. P. Parkin, Y. Lu, X. P. Bian, A. Marley, K. P. Roche, R. A. Altmann, S. A. Rishton, C. Jahnes, T. M. Shaw, and G. Xiao, J. Appl. Phys. **81**, 3741 (1997).

<sup>5</sup>H. Yamane and M. Kobayashi, J. Appl. Phys. **83**, 4862 (1998).

<sup>6</sup>P. Miltényi, M. Gierlings, J. Keller, B. Beschoten, G. Güntherodt, U. Nowak, and K. D. Usadel, Phys. Rev. Lett. **84**, 4224 (2000).

<sup>7</sup>A. P. Malozemoff, Phys. Rev. B **37**, 7673 (1998).

<sup>8</sup>J. Stöhr, A. Scholl, T. J. Regan, S. Anders, J. Lüning, M. R. Scheinfein, H. A. Padmore, and R. L. White, Phys. Rev. Lett. **83**, 1862 (1999).

<sup>9</sup>D. Spanke, V. Solinus, D. Knabben, F. U. Hillebrecht, F. Ciccacci, L. Gregoratti, and M. Marsi, Phys. Rev. B **58**, 5201 (1998).

<sup>10</sup>F. U. Hillebrecht, private communication.

<sup>11</sup>A. Nemoto, Y. Otani, S. G. Kim, K. Fukamichi, O. Kitakami, and Y. Shimada, Appl. Phys. Lett. **74**, 4026 (1999).

<sup>12</sup>Landolt-Börnstein, *Band III/27g* (Springer, Berlin, 1993).

<sup>13</sup>M. T. Hutchings and E. J. Samuelsen, Phys. Rev. B **6**, 3447 (1972).

<sup>14</sup>K. Korosawa, M. Miura, and S. Saito, J. Phys. C **13**, 1521 (1980).

<sup>15</sup>V. Gehanno, P. P. Freitas, A. Veloso, J. Ferreira, B. Almeida, J. B. Sousa, A. Kling, J. C. Soares, and M. F. da Silva, IEEE Trans. Magn. **35**, 4361 (1999).

<sup>16</sup>N. J. Gökemeijer, J. W. Cai, and C. L. Chien, Phys. Rev. B **60**, 3033 (1999).

<sup>17</sup>M. P. Pardavi-Horvath, G. Zheng, G. Vertesy, and A. Magni, IEEE Trans. Magn. **32**, 4469 (1996).

<sup>18</sup>T. Mewes, R. Lopusnik, J. Fassbender, B. Hillebrands, M. Jung, D. Engel, A. Ehresmann, and H. Schmoranzler, Appl. Phys. Lett. **76**, 1057 (2000).