

## Magnetization reversal of $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ layers separated by a nonmagnetic spacer

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We have used polarized neutron reflectometry to individually examine the magnetization reversals of ferromagnetic  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layers separated by a nonmagnetic GaAs spacer layer of varying thickness. For each of the samples studied, the top  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer is adjacent to a Be-doped  $\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}$  capping layer on one side and the GaAs spacer on the other, while the bottom  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer is surrounded by GaAs on either side. For samples with spacer thicknesses of 12 and 6 nm, antiparallel alignment of the two  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer magnetizations was observed at multiple fields, implying that hole doping from the capping layer strongly affects the coercivity of the top  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer but has a weaker effect on the coercivity of the bottom  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer. However, for a spacer thickness of 3 nm, both top and bottom  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layers appear to be equally influenced by the capping layer, as virtually identical coercivities were observed. This behavior is evidence of coupling between the  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layers across the 3 nm GaAs spacer.

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For development of many potential spintronic devices, it is desirable to have semiconductor materials with true long range ferromagnetic (FM) order.  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  has been shown to be just such a material with FM originating from a hole-mediated exchange.<sup>1,2</sup> Interlayer coupling in magnetic multilayer structures is a phenomenon exploited with great utility in numerous device applications,<sup>3</sup> and it is therefore important for spintronic researchers to understand such coupling in  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ -based multilayer devices. Superconducting quantum interference device (SQUID) magnetometry,<sup>4-9</sup> magnetotransport measurements,<sup>4-9</sup> and qualitative analysis of neutron diffraction superlattice peaks<sup>10,11</sup> have been used to indirectly infer evidence of interlayer coupling between separated  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layers. However, the magnetic and structural properties of individual layers in a multilayer structure can be directly obtained through a quantitative analysis of the structure's polarized neutron reflectivity (PNR).<sup>12-15</sup> We have previously reported our use of this technique to precisely determine the structural profiles and temperature ( $T$ ) dependent magnetizations of a series of samples in which two  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layers made to have different (in the absence of any interlayer coupling) Curie temperatures ( $T_C$ ) and coercive fields ( $H_C$ ) are separated by a nonmagnetic GaAs spacer layer of varying thickness.<sup>16</sup> Here, we report on PNR measurements of the same samples performed as function of applied magnetic field ( $H$ ). We observe that for 12 and 6 nm spacers, the two  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layers have very different coercivities, but that for a 3 nm spacer, the coercivities of the two layers are virtually identical.

Three  $1 \times 2 \text{ cm}^2$  rectangular samples were prepared by molecular beam epitaxy on GaAs substrates<sup>17</sup> with the following layer structure (starting at the substrate interface):

- 16 nm bottom  $\text{Ga}_{0.95}\text{Mn}_{0.05}\text{As}$  layer;
- 12, 6, or 3 nm GaAs spacer;
- 8 nm top  $\text{Ga}_{0.95}\text{Mn}_{0.05}\text{As}$  layer; and
- 25 nm  $\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}$  cap doped with Be at a concentration of  $3 \times 10^{20} \text{ cm}^{-3}$ .

Through modulation doping, the Be-doped capping layer is a source of extra holes for the adjacent top  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer.<sup>17,18</sup> The addition of holes affects the hole-mediated FM exchange in  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ ,<sup>1,2</sup> and has been shown to reduce the coercivity.<sup>19</sup> Since the bottom  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer has no adjacent source of extra holes, it will exhibit a  $M(H)$  curve very different from that of the top  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer, unless the two layers are coupled across the spacer.

PNR measurements were conducted using Asterix<sup>14</sup> at the Lujan Neutron Scattering Center of Los Alamos National Laboratory, and the NG-1 Polarized Beam Reflectometer<sup>20</sup> at the NIST Center for Neutron Research. For these measurements, a neutron beam was polarized alternately spin-up (+) and spin-down (−) relative to  $H$  applied along the in-plane magnetic hard [110] (Ref. 16) sample direction, and was incident on the sample. The non-spin-flip specular reflectivities  $R^{++}$  and  $R^{--}$  were measured as a function of wave vector transfer  $Q$ .<sup>21</sup> The data were corrected to account for background, neutron polarization efficiency, beam footprint, and small variations in the critical edge due to slight instrumental misalignment. A sample's depth-dependent nuclear scattering length density  $\rho(z)$ , and the component of the depth-dependent magnetization parallel to  $H$ ,  $M(z)$ , can be deduced

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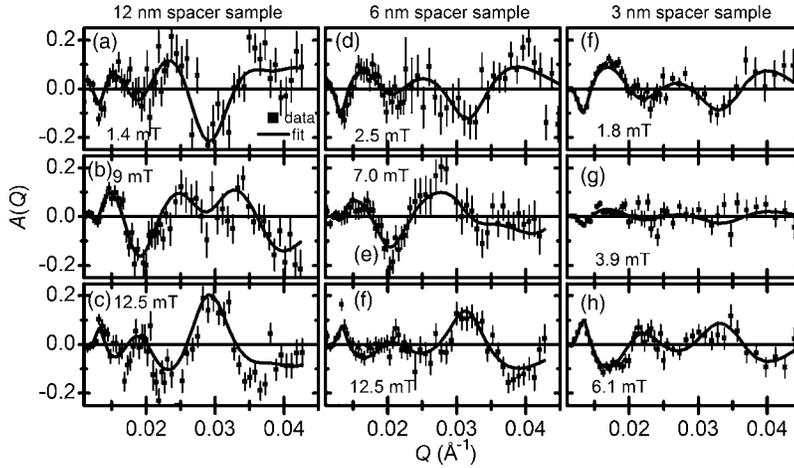


FIG. 1. PNR data (symbols) and fits (lines) shown as spin asymmetry,  $A(Q)$  at selected fields as  $\mu_0 H$  is increased from  $-100$  mT. The 12 nm spacer sample [(a)–(c)], and the 6 nm spacer sample data [(d)–(f)] show evidence of antiparallel alignment of  $M_{\text{top}}$  and  $M_{\text{bot}}$ . The 3 nm spacer sample data [(g)–(i)] shows evidence that  $M_{\text{top}}$  and  $M_{\text{bot}}$  both approach zero at approximately the same field value. Error bars represent  $\pm 1\sigma$ .

by model fitting<sup>22</sup> of  $R^{++}(Q)$  and  $R^{--}(Q)$ .<sup>12–15</sup> In this way, we individually determined the component of the magnetization parallel to  $H$  for the  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer next to the Be-doped AlGaAs cap ( $M_{\text{top}}$ ) and that for the  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer next to the GaAs substrate ( $M_{\text{bot}}$ ) for each of the samples studied. Structural parameters in the models such as layer thickness, interlayer roughness, and Mn concentration  $x$  were determined from the high resolution PNR measurements discussed in Ref. 16. These measurements confirmed the presence of nonmagnetic spacer layers in each of the samples, and revealed that the samples are practically identical in structure and composition except for the thickness of the spacer layer.

$H$ -dependent PNR measurements for each of the samples were conducted after cooling the sample to 6 K in  $\mu_0 H = +100$  mT, lowering to  $\mu_0 H = -100$  mT, and then increasing  $H$  to the desired value. Since the differences between  $R(Q)^{++}$  and  $R(Q)^{--}$  are due to  $M(z)$ , it is intuitive to express the PNR data as spin asymmetry,

$$A(Q) = \frac{R^{++}(Q) - R^{--}(Q)}{R^{++}(Q) + R^{--}(Q)}.$$

Model calculations show that  $A(Q)$  corresponding to both  $M_{\text{top}}$  and  $M_{\text{bot}}$  being negative with respect to  $H$  should only differ from the  $A(Q)$  corresponding to both  $M_{\text{top}}$  and  $M_{\text{bot}}$  being positive with respect to  $H$  by a factor of  $-1$ . Thus, if both top and bottom  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layers reverse in the same way, as  $H$  is increased, the amplitude of the  $A(Q)$  peaks will shrink to zero and then reverse in sign, but no change in the frequency of the oscillations will be observed. However, if  $M_{\text{top}}$  and  $M_{\text{bot}}$  reverse independently of one another, an antiparallel<sup>23</sup> alignment of  $M_{\text{top}}$  and  $M_{\text{bot}}$  is observable as an  $A(Q)$  with nonzero amplitude and a frequency distinct from the frequency corresponding to parallel alignment of  $M_{\text{top}}$  and  $M_{\text{bot}}$ . Figure 1 shows example of  $A(Q)$  data and fits<sup>24</sup> for the samples at selected fields as  $H$  was increased from  $-100$  mT at  $T=5$  K. For the 12 nm spacer sample (panels a–c), the  $A(Q)$  data at 1.4 mT (a) differ from the data at 12.5 mT (c) approximately by a factor of  $-1$ . Model fitting reveals that these two data sets correspond to parallel alignment of both  $M_{\text{top}}$  and  $M_{\text{bot}}$ , positively and negatively aligned with respect to  $H$ , respectively. However, at 9 mT (panel b), the  $A(Q)$  frequency is clearly different from that in

panel a or c. Further, the  $A(Q)$  peak at the lowest  $Q$  has almost zero amplitude, while large amplitudes are observed for higher  $Q$  peaks. Since the smallest  $Q$  corresponds to the largest length scales, this immediately suggests that the average  $M$  of the *entire* sample has approached zero, but that there are local regions of nonzero  $M$ . Indeed, quantitative analysis bears this out, as fitting shows that these data correspond to antiparallel alignment of  $M_{\text{top}}$  and  $M_{\text{bot}}$ . PNR data from the 6 nm spacer sample (panels d–f) reveal similar behavior, with evidence of antiparallel alignment of  $M_{\text{top}}$  and  $M_{\text{bot}}$ . However, the 3 nm spacer sample (panels g–i) is different. Data corresponding to parallel alignment of both  $M_{\text{top}}$  and  $M_{\text{bot}}$ , positive and negative with respect to  $H$ , are observed at 1.8 mT (g) and 6.1 mT (i), respectively. However, no evidence of an antiparallel alignment of  $M_{\text{top}}$  and  $M_{\text{bot}}$  was found at intermediate fields. Instead,  $A(Q)$  with nearly zero amplitude was observed at  $H=3.8$  mT, indicating near zero magnetization parallel to  $H$  for both  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layers.

Fitting results are summarized in Fig. 2, which shows the field dependencies of the individual layer magnetizations for each of the samples. For both the 12 (a) and 6 nm (b) spacer samples, the top  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer is observed to have a significantly smaller  $H_C$  ( $\approx 4$  mT) than the bottom  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layer ( $\approx 10$  mT), as antiparallel alignment of the layers is observed at multiple field values. In principle, antiferromagnetic (AF) interlayer coupling could explain the observed antiparallel alignment, but PNR (Ref. 16) and SQUID (Ref. 9) measurements of the low field  $M(T)$  curves

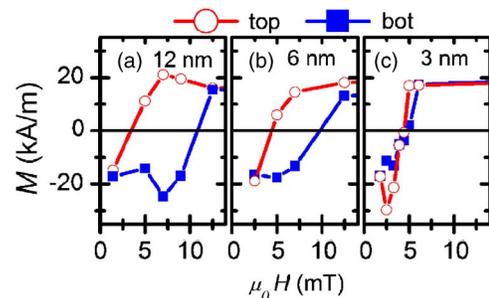


FIG. 2. (Color online) The  $M$  component parallel to  $H$  as a function of  $H$  for the top and bottom  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  layers in (a) the 12, (b) the 6, and (c) the 3 nm spacer samples, as determined from PNR. Solid lines are guides to the eye.

for these samples show that strong AF interlayer coupling is not present.<sup>25</sup> Therefore, our results imply that for spacer thicknesses of 12 and 6 nm, the extra holes supplied by the Be-doped AlGaAs cap strongly affect the magnetic exchange of the top Ga<sub>1-x</sub>Mn<sub>x</sub>As layer, but have a weaker influence over the exchange in the bottom Ga<sub>1-x</sub>Mn<sub>x</sub>As layer. Conversely, the situation is quite different for the 3 nm spacer sample, as *both* the top and bottom Ga<sub>1-x</sub>Mn<sub>x</sub>As layers have  $H_C \approx 4$  mT. Since this value of  $H_C$  is the same as that observed for the top layers of the samples with thicker spacers, this suggests that the influence of the capping layer extends across the 3 nm nonmagnetic GaAs spacer layer, and affects the magnetic exchange of both top and bottom Ga<sub>1-x</sub>Mn<sub>x</sub>As layers.

In Ref. 16, we showed that for these samples, the  $M(T)$  curves of the top and bottom Ga<sub>1-x</sub>Mn<sub>x</sub>As layers become much more similar as the spacer thickness is reduced, regardless of the crystallographic direction along which spins are aligned. The  $M(H)$  results presented here strongly support the primary conclusion drawn from the  $M(T)$  work, namely, that the top and bottom Ga<sub>1-x</sub>Mn<sub>x</sub>As layers are strongly coupled when the spacer between them is 3 nm, and that the coupling weakens as the spacer thickness is increased. That Ga<sub>1-x</sub>Mn<sub>x</sub>As layers can strongly interact with one another across a nonmagnetic spacer layer is a property that may prove important for device applications. While we cannot determine the exact nature of the coupling from our results, we speculate that it could be due to Ruderman–Kittel–Kasuya–Yoshida-like magnetic exchange coupling,<sup>26</sup> and/or an electronic coupling where carrier wave functions in the top Ga<sub>1-x</sub>Mn<sub>x</sub>As layer overlap with those in the bottom Ga<sub>1-x</sub>Mn<sub>x</sub>As layer, resulting in similar hole concentrations for the two layers.<sup>9,27</sup>

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<sup>20</sup>see <http://www.ncnr.nist.gov/instruments/ng1refl/>

<sup>21</sup>Spin-flip scattering, which is sensitive to the component of  $M(z)$  perpendicular to  $H$ , was not measured. Calculations show that due to the small magnitude of  $M$ , any spin-flip scattering from our samples would be weak enough to make such measurements impractical.

<sup>22</sup>P. A. Kienzle, K. V. O'Donovan, J. F. Ankner, N. F. Berk, C. F. Majkrzak (<http://www.ncnr.nist.gov/reflpak>)

<sup>23</sup>Or one layer with zero  $M$  and the other layer with nonzero  $M$ .

<sup>24</sup> $R^{++}$  and  $R^{--}$  were fitted simultaneously, and plotted as  $A$ .

<sup>25</sup>Reference 9 suggests that some *local* AF interlayer coupling may be present in the 6 nm spacer sample but that such coupling is canceled out when considering the sample as whole.

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