Definitive evidence of interlayer coupling between $Ga_{1-x}Mn_xAs$ layers separated by a nonmagnetic spacer

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We have used polarized neutron reflectometry to study the structural and magnetic properties of the individual layers in a series of AlGaAs:Be/$Ga_{1-x}Mn_xAs$/GaAs/$Ga_{1-x}Mn_xAs$ multilayer samples. Structurally, we observe that the samples are virtually identical except for the GaAs spacer thickness (which varies from 3 to 12 nm), and confirm that the spacers contain little or no Mn. Magnetically, we observe that for the sample with the thickest spacer layer, the $Ga_{1-x}Mn_xAs$ layer adjacent to the Be-doped AlGaAs cap has a temperature dependent magnetization very different from that of the other $Ga_{1-x}Mn_xAs$ layer. However, as the spacer layer thickness is reduced, the temperature dependent magnetizations of the two $Ga_{1-x}Mn_xAs$ layers become progressively more similar—a trend we find to be independent of the crystallographic direction along which spins are magnetized. These results definitively show that $Ga_{1-x}Mn_xAs$ layers can couple across a nonmagnetic spacer and that such coupling depends on spacer thickness.

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I. INTRODUCTION

Interlayer coupling in magnetic multilayer structures is a subject of important basic and applied research interest.[1] The most famous example of this is the giant magnetoresistance effect,[2] which is due to interlayer exchange coupling between ferromagnetic (FM) metal layers across a nonmagnetic spacer layer and has been exploited in a multitude of devices.[3] With the recent advent of artificial dilute FM semiconductors such as $Ga_{1-x}Mn_xAs$,[4–7] attention has been given to interlayer coupling in all semiconducting structures.[8] For example, superconducting quantum interference device (SQUID) magnetometry has revealed discrete "steps" in the temperature (T) dependent magnetizations (M) of a series of samples in which two $Ga_{1-x}Mn_xAs$ layers of differing FM transition T were separated by a variable thickness GaAs spacer.[9] These steps were observed to become progressively less pronounced as the GaAs layer thickness was reduced, suggesting that the two $Ga_{1-x}Mn_xAs$ layers were strongly coupled when close to one another, but less so when more distant. Additionally, other SQUID measurements,[10–13] magnetotransport measurements,[9–11,13] and qualitative analysis of neutron diffraction superlattice peaks[14,15] have yielded some evidence of interlayer coupling in $Ga_{1-x}Mn_xAs$ based multilayer structures. However, such evidence is indirect, having been obtained through techniques which can only infer the behavior of individual layers from the collective behavior of the entire multilayer structure. Conversely, a quantitative analysis of the structure's polarized neutron reflectivity (PNR) can resolve the M of each layer in such a structure.[16–18] Using this technique, we have gone beyond previous studies and have directly measured $M(T)$ for the individual layers in a series of $Ga_{1-x}Mn_xAs$ based multilayer structures. Our results unambiguously show that $Ga_{1-x}Mn_xAs$ layers can strongly couple across a nonmagnetic spacer layer, and that such coupling is dependent on the spacer thickness.

II. EXPERIMENTAL RESULTS

PNR measurements were conducted using the NG-1 Reflectometer at the NIST Center for Neutron Research. A neutron beam of wavelength 4.75 Å was polarized alternately spin up (+) and spin down (−) relative to a magnetic field H applied in the plane of the sample. This beam was incident on the sample, and the non-spin-flip specular reflectivities R^{++} and R^{--} were measured as a function of wave vector transfer Q.[19] Standard corrections were applied to the data to correct for background, neutron polarization efficiencies, and footprint of the incident beam. A sample's depth-dependent nuclear scattering length density $\rho(z)$ and magnetization component parallel to H, $M(z)$, can be deduced by model fitting of $R^{++}(Q)$ and $R^{--}(Q)$.[16–18] Employing the REFLPAK software suite,[20] we used such model fitting to determine the thickness, Mn concentration x,[21,22] and M for the individual layers in each of the samples studied.[23]

Three 1×2 cm^2 rectangular samples were prepared by molecular beam epitaxy on GaAs substrates[24] with the following layer structure (starting at the substrate interface):

(1) 16 ± 2 nm bottom layer of FM $x=0.05\pm 0.015$ $Ga_{1-x}Mn_xAs$,

(2) variable thickness nonmagnetic GaAs spacer,

(3) 8 ± 2 nm top layer of FM $x=0.05\pm 0.015$ $Ga_{1-x}Mn_xAs$,

(4) 25 ± 1.5 nm $Al_{0.25}Ga_{0.75}As$ cap doped with Be at a concentration of 3×10^{20} cm^{-3}.

All three samples were grown under the same conditions as part of the same run. We observe that nominally[25] the three samples are structurally identical except for the spacer thickness, which are 12 ± 1, 6 ± 1, and 3 ± 1 nm, respectively.[26,27] Figure 1 shows the fitted PNR data taken out to highest Q and, therefore, used to determine the structural parameters of the samples. These data were taken at H and T values corresponding to conditions where the magnetizations of the top (adjacent to the Be-doped AlGaAs cap) and bot-

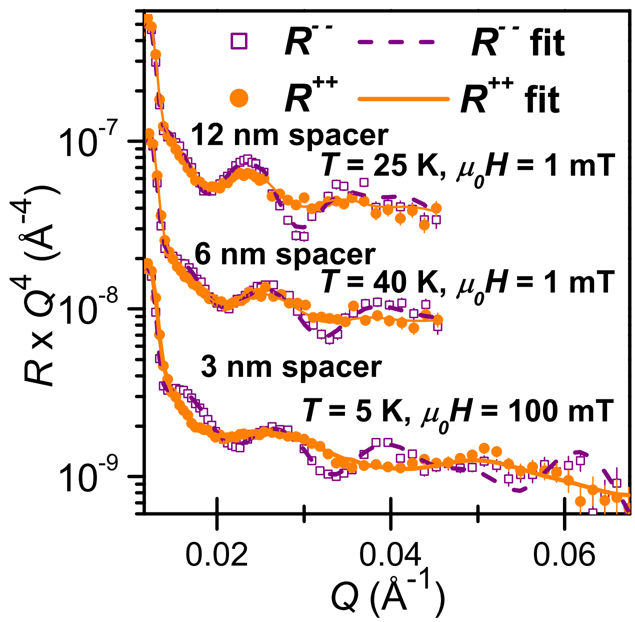

FIG. 1. (Color online) PNR data and fits used to determine the layer thicknesses and compositions for the samples. The data are vertically offset and multiplied by Q^4 for clarity. Error bars represent $\pm 1\sigma$.

tom $Ga_{1-x}Mn_xAs$ layers (M_{top} and M_{bot}) are similar to each other and nonzero. For all three samples, fits to the PNR data are extremely sensitive to the presence of a spacer layer with greatly reduced x (consistent with 0) and greatly reduced M (also consistent with 0) in the model. Fitting error associated with these parameters suggests that the spacer has at most approximately ten times smaller x and approximately four times smaller M than the adjacent $Ga_{1-x}Mn_xAs$ layers. Models that treat the $Ga_{1-x}Mn_xAs/GaAs/Ga_{1-x}Mn_xAs$ structure as a single layer of uniform x and/or M result in substantially worse fits to the data. This confirms that little or no Mn is present in the spacers of these samples.

Since the FM exchange in $Ga_{1-x}Mn_xAs$ is hole mediated,[6,7,28] the FM transition T of a $Ga_{1-x}Mn_xAs$ layer can be increased by placing it adjacent to a Be-doped AlGaAs layer, which is a source of holes.[22,29] For each of our samples, only the top $Ga_{1-x}Mn_xAs$ layer is adjacent to such a hole source. Therefore, the top and bottom $Ga_{1-x}Mn_xAs$ layers will exhibit very different $M(T)$ curves—unless the two layers interact across the spacer. In order to test for such coupling and examine how it changes with spacer thickness, we used PNR to measure $M(T)$ of the individual layers of each sample at a field of 1 mT.[30] To examine the effects of anisotropy, these measurements were conducted for H along each of the in-plane crystallographic directions: The [100], a [110] found to be magnetically hard, and a $[1\bar{1}0]$ found to be magnetically easy. Because the densities of the samples do not change appreciably over the T range examined, T-dependent changes in a sample's PNR can be solely attributed to magnetic changes in the sample. Since the magnetic properties of the sample are manifested in the differences between $R(Q)^{++}$ and $R(Q)^{--}$, it is intuitive to express the PNR data as spin asymmetry:

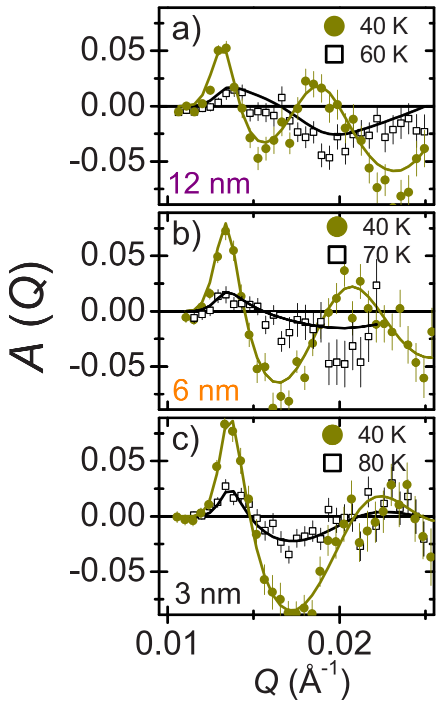

FIG. 2. (Color online) PNR data and fits (solid lines) expressed as spin asymmetry for the (a) 12 nm spacer sample, (b) 6 nm spacer sample, and (c) 3 nm sample, taken with $\mu_0 H = 1$ mT along the hard [110] direction. Error bars represent $\pm 1\sigma$.

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

Model calculations show that the frequency of the $A(Q)$ oscillations is strongly dependent on the *total* magnetized thickness of the entire multilayer structure. Thus, the $A(Q)$ frequency exhibited when M_{top} and M_{bot} are similar is distinct from the frequency exhibited when M_{top} and M_{bot} are very different. The amplitude of the peaks is largely dependent on the total M of the sample. Therefore, the sample's $A(Q)$ falls into one of three general categories:

(1) A high frequency oscillation corresponding to low T, where $M_{top} \approx M_{bot}$.

(2) A lower frequency oscillation corresponding to $M_{top} \neq 0$ and $M_{bot} \approx 0$.

(3) $A(Q) = 0$, corresponding to $M_{top} = M_{bot} = 0$.

As an example, Fig. 2 shows $A(Q)$ data at selected T for each of the samples, measured with $\mu_0 H = 1$ mT along the hard [110] direction. The solid lines through the data points are derived from fits to $R^{++}(Q)$ and $R^{--}(Q)$. For the 12 nm spacer sample in Fig. 2(a), clearly different $A(Q)$ frequencies are exhibited for the two T, indicating that the total magnetized thickness of the sample changes between 40 and 60 K. Figure 2(b) indicates a similar transition between 40 and 70 K for the 6 nm spacer sample. In contrast, Fig. 2(c) shows no change in frequency for the 3 nm spacer sample between 40 and 80 K, indicating that the total magnetized thickness does not change over this T range. Figure 2 presents a compelling qualitative argument that the $M(T)_{top}$ and

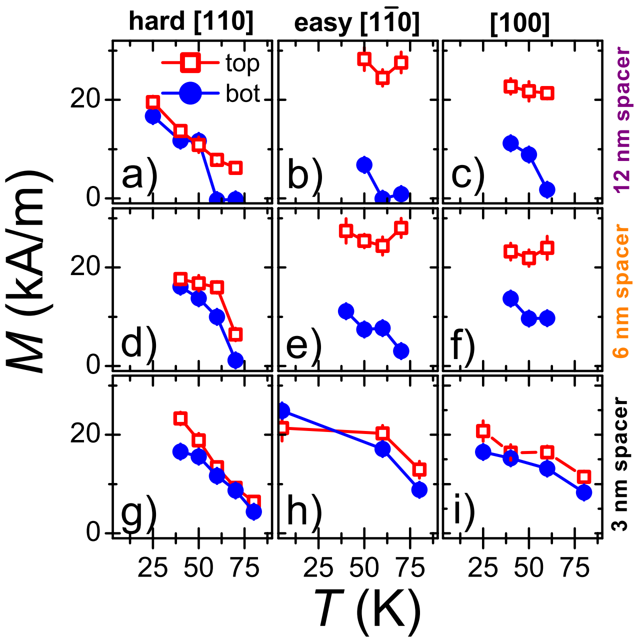

FIG. 3. (Color online) $M(T)$ for the individual top and bottom $Ga_{1-x}Mn_xAs$ layers for the three samples (rows), for H along each of the three in-plane crystallographic directions (columns). Solid lines are guides for the eye. Error bars represent $\pm 1\sigma$.

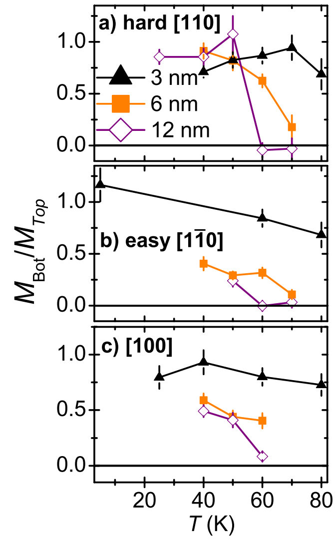

FIG. 4. (Color online) Ratio of the bottom and top $Ga_{1-x}Mn_xAs$ layer magnetizations for each of the samples for H along (a) the hard [110], (b) the easy [1$\bar{1}$0], and (c) the [100]. Solid lines are guides for the eye. Error bars represent $\pm 1\sigma$.

$M(T)_{bot}$ become more similar as the spacer layer thickness is reduced.

Quantitative results obtained from fits to the PNR data are presented in Fig. 3, which shows the full $M(T)_{top}$ and $M(T)_{bot}$ curves for H along each of the in-plane crystal directions. We note that the average sample magnetizations corresponding to the curves in Fig. 3 (Ref. 31) agree well with those obtained from T-dependent SQUID measurements of the same samples, which are presented elsewhere.[32] While PNR is a direct probe of the total sample M in and of itself, this agreement with SQUID further confirms that the models used to fit the PNR data are reasonable. Effects of T-dependent anisotropy are evident for the 12 nm spacer sample and the 6 nm spacer sample in Fig. 3, as the relationship between $M(T)_{top}$ and $M(T)_{bot}$ for the hard [110] direction is significantly different than it is for the easy [1$\bar{1}$0] or the [100] direction. Such effects are not so apparent for the 3 nm spacer sample, as $M(T)_{top}$ and $M(T)_{bot}$ look comparatively similar to one another in all three directions. The differences among the samples are easily seen in Fig. 4, which shows $M(T)_{bot}/M(T)_{top}$ for each one. The T at which this ratio goes to zero indicates the vanishing of M_{bot}. For all three directions, $M(T)_{bot}/M(T)_{top}$ is nonzero below 50 K for the 12 nm sample, below 60 K for the 6 nm sample, and below 80 K for the 3 nm sample. Figure 4 clearly illustrates the primary finding of this paper: Regardless of magnetization direction, $M(T)_{top}$ and $M(T)_{bot}$ become progressively more similar to each other as the spacer thickness between them is reduced.

III. DISCUSSION AND CONCLUSIONS

Since the three samples were grown under nominally identical conditions, and are observed to be virtually the same except for spacer thickness, this behavior is definitive evidence of a coupling between the $Ga_{1-x}Mn_xAs$ layers that varies in strength as the distance between them increases.[33] While it has been shown that small quantities of Mn can diffuse as far as 1.5 nm into the GaAs layer of $Ga_{1-x}Mn_xAs$/GaAs (Ref. 34) structures, we conclude that the observed coupling cannot be mediated by a "magnetic short" resulting from unintended FM order in the spacer layer. As discussed previously, at 100 mT and 5 K (conditions approaching magnetic saturation of $Ga_{1-x}Mn_xAs$), we observe for the 3 nm spacer sample that the spacer has greatly reduced x and M (i.e., consistent with zero) compared to the surrounding $Ga_{1-x}Mn_xAs$ layers. Figure 4 shows that the coupling between the $Ga_{1-x}Mn_xAs$ layers in this sample persists up to at least $T=80$ K. Therefore, for FM bridges or shorts in the spacer to be responsible for the interlayer coupling, FM order in a layer with no observable magnetic dopant or low-T M would also have to persist above 80 K. Considering that M for the bottom $Ga_{1-x}Mn_xAs$ layer of the 12 nm spacer sample (which has $x=0.05$ and a sizable M at low T) disappears below 60 K, such a scenario is totally implausible. Thus, if Mn is present in the spacer layers, it can be considered magnetically inactive.

We instead propose that itinerant holes are responsible for the observed spacer-dependent interlayer coupling. One possibility is that interlayer hopping of free holes results in a Ruderman-Kittel-Kasuya-Yosida (RKKY)-like FM exchange coupling of the two $Ga_{1-x}Mn_xAs$ layers that becomes stronger as the spacer thickness is reduced.[8] Another possibility is that the two $Ga_{1-x}Mn_xAs$ layers are electronically coupled by holes.[32] As the spacer thickness is reduced, the wave functions of holes in the top $Ga_{1-x}Mn_xAs$ layer may begin to significantly overlap the wave functions of holes in the bottom $Ga_{1-x}Mn_xAs$ layer.[35] Such an overlap could make it easier for carriers in the hole-rich top $Ga_{1-x}Mn_xAs$ layer to overcome the potential barrier of the GaAs spacer and drift into the bottom $Ga_{1-x}Mn_xAs$ layer, resulting in a more equal distribution of holes and, thereby, more similar FM properties in the two $Ga_{1-x}Mn_xAs$ layers.[32] Either of these coupling mechanisms, or a combination of the two, could explain our results.

In conclusion, we have confirmed what was suggested by experiments in Refs. 9–15, that $Ga_{1-x}Mn_xAs$ layers can strongly couple across a nonmagnetic spacer layer. Additionally, we observe evidence of coupling across a spacer 6 nm thick, a factor of 3 greater than the thickness for which it has been theoretically predicted that RKKY-like FM coupling should effectively disappear.[8] These results show the robustness of the carrier-mediated interaction between separated $Ga_{1-x}Mn_xAs$ layers, a quality that may prove beneficial for device applications.

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ruled out. However, it is exceedingly unlikely that such variations could cause the spacer-dependent trends that we report on in this paper, considering that the samples were all grown in the same run, under nominally identical growth conditions, and PNR reveals no evidence of structural differences among the samples.

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