Interlayer exchange in (Ga,Mn)As/(Al,Ga)As/(Ga,Mn)As semiconducting ferromagnet/nonmagnet/ferromagnet trilayer structures

N. Akiba, F. Matsukura, A. Shen, Y. Ohno, and H. Ohno

Laboratory for Electronic Intelligent Systems, Research Institute of Electrical Communication, Tohoku University, Sendai 980-8577, Japan

A. Oiwa, S. Katsumoto, and Y. Iye

Institute for Solid State Physics, University of Tokyo, Tokyo 106-8066, Japan

(Received 19 May 1998; accepted for publication 7 August 1998)

Magnetic properties of all-semiconductor (Ga,Mn)As/(Al,Ga)As/(Ga,Mn)As trilayer structures are studied. The interactions between the two ferromagnetic (Ga,Mn)As layers are investigated by magnetotransport measurements in a number of samples with different GaAs thickness or with different Al content in the intermediary nonmagnetic (Al,Ga)As layer. The results indicate that carriers present in the nonmagnetic layer mediate the coupling between the two ferromagnetic layers. © 1998 American Institute of Physics. [S0003-6951(98)01441-7]

Ferromagnet/nonmagnet multilayers are currently of great interest and extensively studied both experimentally and theoretically.1 In metallic systems such as Fe/Cr multilayers, the coupling between ferromagnetic layers (Fe) separated by nonmagnetic metal layers (Cr) was found to oscillate as a function of the thickness of the nonmagnetic layers, due basically to the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction; antiferromagnetic coupling in such metal multilayer systems was shown to lead to giant magnetoresistance (GMR).2 When nonmagnetic metal was replaced by an insulator, spin-dependent tunneling between the ferromagnetic metal layers gave rise to tunneling magnetoresistance; its basic physics as well as its application to magnetic recording technology are being widely studied.3 When the semiconductor was used for the intermediary nonmagnetic layer, the strength of interlayer coupling became a function of external parameters such as temperature4 and illumination.5 So far, to the best of our knowledge there has been no study on magnetic coupling in the all-semiconductor ferromagnet/nonmagnet multilayer system because of the lack of a suitable ferromagnetic semiconductor that can be used to form multilayer structures by semiconductors alone. On the other hand, there is a number of advantages of the all-semiconductor system over metallic systems. To give a few examples, the higher resistivity of semiconductors makes it easier to work on vertical transport of thin samples, the longer Fermi wavelength than that of metal together with established submicron processing technology in semiconductors allows easier observation of quantum mechanical effects, and the interlayer exchange may be controlled not only by illumination but also by electric fields if the coupling is mediated by carriers in the intermediary semiconducting layer.

The III–V based diluted magnetic semiconductor (Ga,Mn)As is an ideal candidate to study interlayer magnetic coupling in all-semiconductor ferromagnet/nonmagnet multilayers, since it is ferromagnetic, it can be grown by low-temperature molecular beam epitaxy (LT-MBE) on GaAs substrates, and thus, is compatible with the (Al,Ga)As/GaAs heterostructure system.6 The ferromagnetism of (Ga,Mn)As has been explained in terms of the RKKY interaction mediated by holes, which are introduced by Mn of (Ga,Mn)As.7 (Ga,Mn)As/GaAs superlattices have been realized and shown to have high crystal perfection and good interface quality, together with ferromagnetic order at low temperature.8 In this letter, we report the magnetic properties of all-semiconductor ferromagnet/nonmagnet/ferromagnet trilayer structures based on (Ga,Mn)As. We examine, by the use of magnetotransport measurements, the presence of a carrier mediated interaction between the (Ga,Mn)As layers separated by a nonmagnetic (Al,Ga)As or GaAs layer.

The trilayer structures were grown by LT-MBE (substrate temperature $T_{\text{sub}} = 250^\circ$C) on a 150 nm (Al$_{0.3}$Ga$_{0.7}$)As buffer layer, grown at 560 °C on semi-insulating GaAs(001) substrates. Two sets of trilayer structures were prepared. The structure of one set consists of a top 30 nm (Ga,Mn)As layer (Mn composition, $x_{\text{Mn}} = 0.04$) and a bottom 30 nm (Ga, Mn)As layer ($x_{\text{Mn}} = 0.02$), separated by a nonmagnetic GaAs layer with thickness $d_{\text{GaAs}}$ ranging from 0 to 106 monolayers (MLs). The other set is identical in structure except for the nonmagnetic intermediary layer, where 10 MLs of (Al,Ga)As with Al composition of $x_{\text{Al}} = 0.16$ or 0.29 was used. The different value of $x_{\text{Mn}}$ for the two (Ga,Mn)As layers was introduced intentionally to distinguish the magnetic behavior of the two layers. Single layers of 30 nm (Ga,Mn)As on (Al$_{0.3}$Ga$_{0.7}$)As with $x_{\text{Mn}} = 0.02$ and 0.04 were also prepared and measured for reference. From the measurements of the reference layers, the ferromagnetic transition temperature $T_c$ of (Ga,Mn)As with $x_{\text{Mn}} = 0.02$ and 0.04 is determined to be at around $T_c = 40$ and 80 K, respectively.

Magnetotransport properties of the trilayer structures were measured with a standard dc setup using Hall bar geometry. The sheet resistance $R_{\text{sheet}}$ and the Hall resistance $R_{\text{Hall}}$ were measured simultaneously in magnetic fields $B$ perpendicular to the plane (the direction of the hard axis). Since $R_{\text{Hall}}/R_{\text{sheet}}$ of (Ga,Mn)As is known to be approximately proportional to the magnetization $M$ of the film (because of the...
dominant contribution of the anomalous Hall effect, which is proportional to \( M \) perpendicular to the plane), the measured \( R_{Hall}/R_{sheet} - B \) curves reflect the \( M-B \) curve of the sample. The \( R_{sheet} \) and \( R_{Hall} \) of the two reference layers (\( x_{Mn} = 0.02 \) and \( x_{Mn} = 0.04 \)) measured at 17 K (not shown) revealed that the saturation field \( B_{sat} \), where \( M \) saturates, was lower for the \( x_{Mn} = 0.02 \) layer than the \( x_{Mn} = 0.04 \) layer. \( R_{sheet} \) of the \( x_{Mn} = 0.02 \) layer was about three orders of magnitude higher than that of the \( x_{Mn} = 0.04 \) layer. This resistivity difference allows one to probe only the properties of the \( x_{Mn} = 0.04 \) layer in the trilayer structures by the magnetotransport measurements.

Figure 1(a) shows \( R_{Hall}/R_{sheet} - B \) curves of the trilayer structures with \( n \) ML of the intermediary nonmagnetic GaAs layer at 2 K. As the thickness of the GaAs layer decreases (from \( n = 106 \) to 0), \( B_{sat} \) (of the top layer) decreases. The reduction of \( B_{sat} \) shows stronger coupling between the two magnetic layers, since the bottom (Ga,Mn)As layer (\( x_{Mn} = 0.02 \)) has lower \( B_{sat} \) than that of the top layer. The results of the \( B \) dependence of \( R_{Hall}/R_{sheet} \) of the two (Ga,Mn)As/10 MLs \((Al,Ga)As/(Ga,Mn)As\) trilayer structures at 2 K are shown in Fig. 1(b) (\( x_{Al} = 0.16 \) and 0.29). This time the nonmagnetic \((Al,Ga)As\) layer acts as a potential barrier in the valence band; the valence-band barrier between GaAs and \((Al,Ga)As\) is 0.55\(x_{Al}\) eV. As can be seen in Fig. 1(b), \( B_{sat} \) of \( x_{Al} = 0.29 \) is higher than that of \( x_{Al} = 0.16 \), showing weakening of coupling with increasing the barrier height in the valence band.

To probe further the nature of the interlayer coupling, magnetization of the samples were measured by a superconducting quantum interference device (SQUID) magnetometer at 5 K with magnetic-field \( B \) applied parallel to the sample plane (the direction of the easy axis). \( M-B \) curves of single (Ga,Mn)As layers with \( x_{Mn} = 0.02 \) and 0.04 (not shown) exhibited a clear hysteresis loop and the \( x_{Mn} = 0.02 \) layer showed a higher coercive field \( B_c \) (\( \sim 0.015 \) T) and a softer

\[ M-B \] curve than those of the \( x_{Mn} = 0.04 \) layer (\( B_c \sim 0.009 \) T). The height of hysteresis loop for \( x_{Mn} = 0.02 \) was half of that of \( x_{Mn} = 0.04 \), which is in agreement with the expected ratio from the nominal \( x_{Mn} \) of the samples. The \( M-B \) curves (\( B \) applied in plane) of (Ga,Mn)As/(Al,Ga)As/(Ga,Mn)As trilayer structures measured at 5 K are shown in Fig. 2. Two steps in the magnetization process were observed for the \( x_{Al} = 0.29 \) sample. Since this \( M-B \) curve can be reproduced by a simple addition of the two individual \( M-B \) curves measured separately on single (Ga,Mn)As layers, it indicates that there is no appreciable magnetic coupling between the two (Ga,Mn)As layers. On the other hand, such a double step was not observed in the \( M-B \) curve of the sample with \( x_{Al} = 0.16 \). This shows that the two (Ga,Mn)As layers are magnetically coupled, and as a result of coupling, the magnetization reversal of the two (Ga,Mn)As layers occurs at the same time. In the \( M-B \) curves of (Ga,Mn)As/(Al,Ga)As/(Ga,Mn)As trilayer structures (not shown), such double-step structures were not clearly resolved, because the hysteresis loop was still open at the high field region due, most probably, to the presence of a spin-glass-like component, which was observed in the reference (single-layer) samples as well. The origin of this component is not fully understood at the moment.

The coupling between the two (Ga,Mn)As layers can be quantified in the following way. Total energy per unit volume of a single magnetic layer can be expressed as

\[ E = K \cos^2 \theta_K (1 - \alpha \cos^2 \theta_K) - (MB/\mu_0) \cos \theta - J \cos \theta \]

(1)

where the first term is the anisotropy energy, the next the Zeeman energy, and the third the coupling energy between the two (Ga,Mn)As layers [\( K \) is the anisotropy energy density, \( \theta_K \) the angle between \( M \) and the hard axis, \( \theta \) the angle between \( M \) and \( B \), \( \theta_j \) the angle between \( M \) of top (Ga,Mn)As and \( M \) of the bottom (Ga,Mn)As, \( \alpha \sim -0.035 \) an anisotropy constant introduced to describe the magnetization jump around \( B \sim 0.2 \) T, \( \mu_0 \) the magnetic permeability of vacuum, and \( J \) the coupling energy density]. The minimum condition of Eq. (1) gives the magnetization of the structure along \( B \). Because \( B_{sat} \) of the \( x_{Mn} = 0.02 \) reference layer is lower than
the interaction of two thin films. For samples with nonmagnetic GaAs layers, we assume \( J \) becomes weaker. \( J \) becomes higher, holes in the nonmagnetic layer decrease and the magnetic interaction becomes stronger.

\[ \text{FIG. 3. (a) Thickness and (b) Al content dependence of coupling energy between two (Ga,Mn)As layers.} \]

\[ B_{\text{sat}} \text{ of } x_{\text{Mn}} = 0.04, \text{we may assume that the magnetization of the bottom } x_{\text{Mn}} = 0.02 \text{ layer is along } B \text{ and fully saturated when the top } x_{\text{Mn}} = 0.04 \text{ layer starts its reversal. We can then set } \theta = \theta_{K} = \theta_{J}, \text{ since } B, M \text{ of the bottom layer, and the direction of the hard axis are now all perpendicular to the sample plane. } K \text{ is the same for all the } x_{\text{Mn}} = 0.04 \text{ layers, because all the } x_{\text{Mn}} = 0.04 \text{ layers have the same thickness and the same geometry.} \]

For analysis of samples with nonmagnetic GaAs layers, we assume \( J = 0 \) for the 106 ML sample, which we believe is reasonable because 30 nm is thick enough to cut off any magnetic interaction. Static dipole interaction (long range) does not play an important role, since we are dealing with the interaction of two thin films. For samples with (Al,Ga)As layers, we assume \( J = 0 \) for the \( x_{\text{Al}} = 0.29 \) sample since the two magnetic layers are shown to be decoupled by the magnetization measurements. Since nonzero \( J \) alters the magnetization process, we can deduce \( J \) from the difference in the magnetization curves.

Figure 3 shows \( J \) as a function of thickness or composition of the intermediary layer. \( J \) is always ferromagnetic because \( B_{\text{sat}} \) is always reduced when the interaction sets in. The magnetic coupling of Fig. 3 can be qualitatively explained by the presence of the carrier-mediated magnetic interaction (the RKKY interaction). Solutions of Poisson’s equation show that there are almost no holes present in the GaAs layer when the spacer layer thickness is 30 nm (106 MLs) and the hole concentration in the GaAs layer increases as the thickness of the intermediary layer decreases. The valence-band barrier height produced by (Al,Ga)As becomes over 150 meV for \( x_{\text{Al}} = 0.29 \), which is high enough to allow almost no carriers in the (Al,Ga)As layer. The barrier produced by (Al,Ga)As with \( x_{\text{Al}} = 0.16 \) is about 80 meV, now allowing holes to be present in the (Al,Ga)As layer. Our results thus show that the magnetic coupling becomes stronger as the hole concentration in the nonmagnetic layer increases. The hole concentration in the (Ga,Mn)As layers are low compared to metals, of the order of \( 10^{20} \text{ cm}^{-3} \) at most, and the carrier concentration in the nonmagnetic layer should be even lower than that of the (Ga,Mn)As layer. This low carrier concentration results in a long-range ferromagnetic interaction. When the interaction changes its sign due to the oscillatory nature of the RKKY interaction, the magnitude of the interaction is too low to be observed.

In conclusion, we have shown that the magnetic coupling between two ferromagnetic layers separated by a nonmagnetic layer in all-semiconductor (Ga,Mn)As/(Al,Ga)As/(Ga,Mn)As trilayer structures is a function of the thickness of the Al content of the nonmagnetic layer. The magnetic coupling can qualitatively be explained by the magnetic interaction mediated by holes in the nonmagnetic layer. These results suggest the possibility of new devices in which the magnetic interlayer coupling is controlled by external parameters such as electric fields or optical excitations, which are widely used to control the carrier concentration of semiconductor layers.

The authors thank Professor J. Inoue of Nagoya University for useful discussions. This work was supported by a Grant-in-Aid for Science Research on Priority Area “Spin Controlled Semiconductor Nanostructures” (Grant No. 09244103) from the Ministry of Education, Science, Sports and Culture, Japan, and by the “Research for the Future” Program (Grant No. JSPS-RFTF97P00202) from the Japan Society for the Promotion of Science.