PHYSICAL REVIEW B **71**, 201303(R) (2005)

Interlayer exchange coupling in (Ga,Mn)As-based superlattices

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(Received 15 February 2005; published 11 May 2005)

The interlayer coupling between (Ga,Mn)As ferromagnetic layers in all-semiconductor superlattices is studied theoretically within a tight-binding model, which takes into account the crystal, band and magnetic structure of the constituent superlattice components. It is shown that the mechanism originally introduced to describe the spin correlations in antiferromagnetic EuTe/PbTe superlattices, explains the experimental results observed in ferromagnetic semiconductor structures, i.e., both the antiferromagnetic coupling between ferromagnetic layers in IV-VI (EuS/PbS and EuS/YbSe) superlattices as well as the ferromagnetic interlayer coupling in III-V [(Ga,Mn)As/GaAs] multilayer structures. The model also allows prediction of (Ga,Mn)As-based structures, in which an antiferromagnetic interlayer coupling could be expected.

DOI: 10.1103/PhysRevB.71.201303 PACS number(s): 75.50.Pp, 68.65.Cd

Interlayer exchange coupling (IEC)—the phenomenon, which was shown to be responsible for the giant magnetoresistance effect,[1] and which already led to many applications of magnetic metallic thin film structures[2]—was discovered in late 1980s. Since the first report on correlated magnetization vectors in Fe/Cr/Fe trilayers,[3] IEC was observed in a variety of structures composed of metallic ferromagnetic (FM) layers separated by nonmagnetic, metallic, or insulating spacer layers. The attempts to explain this phenomenon were summarized in Ref. 4, where it was shown that IEC can be ascribed to the spin-dependent changes of the density of states resulting from the quantum interference of conduction-electron waves.

Although the FM and the metallic character of magnetic layers were considered as inherent elements of the IEC effect, in 1995 the interlayer spin correlations between antiferromagnetic (AFM) layers in all-semiconductor superlattices (SL) were reported.[5] Next, such coupling was also identified in semiconductor multilayer structures with FM, (Ga,Mn)As (Ref. 6), and EuS (Ref. 7), layers. In addition to their basic science significance, these discoveries were important because the all-semiconductor structures offer the possibility to overcome the limitations brought about by the technological incompatibility of FM metals and semiconductors. Moreover, their properties can be easily controlled by temperature, light, or external electric fields. From this applicational point of view, the most interesting was the discovery of AFM coupling between FM layers in EuS/PbS SL (Ref. 7). In these structures, however, the effect takes place only at very low temperatures—bulk EuS is a classical Heisenberg ferromagnet with the Curie temperature 16.6 K (Ref. 8). In (Ga,Mn)As-based FM structures, where a higher critical temperature can be achieved, unfortunately only FM IEC was observed.[6,9–12]

To explain the spin correlations observed in the AFM EuTe/PbTe and FM EuS/PbS SL, a model was proposed,[13] in which the significant role of the valence-band electrons in IEC in all-semiconductor magnetic/nonmagnetic layer structures was put in evidence. In Ref. 13 it has been proven that quantum interference between the spin-dependent perturbations in successive barriers, as proposed by Bruno,[4] is an effective mechanism for magnetic long-range correlations also when there are no free carriers in the system. The IEC mediated by valence-band electrons, calculated within this model, correlates antiferromagnetically the spins at the two interfaces bordering each nonmagnetic layer of the SL. Such spin-spin interactions lead, in agreement with the experimental findings, to zero net magnetic moment in the case of AFM EuTe/PbTe SL (Ref. 14) and to an AFM coupling between successive FM EuS layers in EuS/PbS SL (Refs. 7 and 15). The strength of the obtained IEC decreases rapidly (exponentially) with the distance between the spins, i.e., with the thickness of the nonmagnetic spacer layer and practically does not depend on the thickness of magnetic layers. In Refs. 15 and 16 a careful analysis of the experimental results, in particular of the temperature and magnetic-field dependence of the magnetization, led the authors to the conclusion that such IEC describes properly all the neutron scattering and magnetic observations in EuS/PbS structures with ultrathin (ca. 1.2-nm-thick) PbS spacers. The traces of the coupling observed by neutron scattering in samples with relatively thick spacers were ascribed, however, to the weak but slowly decaying contribution from the dipolar interactions.[7,16,17]

In the (Ga,Mn)As-based semiconductor ferromagnetic/nonmagnetic systems interlayer coupling of opposite FM sign was observed—by magnetic measurements,[6,9,12] neutron diffraction,[11] and polarized neutron reflectometry.[10] These structures differ from the previously considered EuS/PbS multilayers by many aspects, which all can affect the IEC. First of all, in contrast to the simple rocksalt crystal structure of EuS-based SL, they crystallize in a zinc-blende structure. Moreover, PbS is a narrow gap, whereas EuS is a wide gap semiconductor. In EuS/PbS SL the spacer layers form deep wells in the energy structure of the multilayer. Here, the band structures of the magnetic [(Ga,Mn)As] and nonmagnetic [GaAs, (Al,Ga)As] materials are either very similar or the spacer layers introduce potential barriers for the carriers. It should be noted, however, that in EuS-based structures the wider energy gap of the spacer material does not lead to different character of IEC, but results only in a reduction of the coupling strength and range. This was shown by theoretical studies of the coupling between EuS layers separated by YbSe and SrS insulators[18] and confirmed by neutron

reflectivity experiments in EuS/YbSe SL (Ref. 19). Finally, (Ga,Mn)As is not a magnetic, but diluted magnetic semiconductor (DMS). In this ternary alloy a small, randomly distributed fraction of the Ga cations is substituted by magnetic Mn ions. The spin splittings are smaller than in EuS; the ferromagnetism is carrier induced[20] and requires a considerable amount of free holes in the valence band of the FM (Ga,Mn)As.

In Refs. 6 and 9 the observed much weaker IEC in samples with high (30%) Al content in the (Al,Ga)As spacer led the authors to the conclusion that the coupling between the FM layers is mediated by the carriers in the nonmagnetic layer. Recently, it was also shown that introducing extra holes by Be doping of the GaAs spacer increases the interlayer coupling.[12] To explain the spin correlations between (Ga,Mn)As layers the RKKY mechanism and the models tailored for metallic systems were invoked.[21,22] In this paper, in order to describe the spin-dependent bandstructure effects that can lead to IEC in (Ga,Mn)As-based semiconductor SL, we built a tight-binding model in the spirit of the approach used before for IV-VI semiconductor magnetic multilayers.[13] The model was applied to the SL consisting of alternating m monolayers of (Ga,Mn)As, with the Mn content 4% or 6%, and n monolayers of GaAs, (Al,Ga)As, or GaAs:Be, i.e., to the structures studied experimentally.

In order to construct the empirical tight-binding Hamiltonian matrix for the SL, one has to describe first the constituent materials, select the set of atomic orbitals for every type of involved ions, and specify the range of the ion-ion interactions. In the following, we assume that the proper description of SL band structure is reached when the Hamiltonian reproduces in the $n=0$ and $m=0$ limits the band structures of the constituent magnetic and nonmagnetic materials, respectively. Bulk GaAs is tetrahedrally coordinated cubic material in which each cation (anion) is surrounded by four anion (cation) nearest neighbors (NN) along the [1, 1, 1], $[1,-1,-1]$, $[-1,1,-1]$, and $[-1,-1,1]$ directions, at the distances $a\sqrt{3}/4$ (where $a=5.653$ Å is the lattice constant). GaAs is a nonmagnetic, direct gap semiconductor with the valence-band maximum at the center of the Brillouin zone. The top of the valence band is formed by two twofold-degenerate p bands. The third p band is separated from the two by spin-orbit splitting, $\Delta_{so}=0.34$ eV. The band structure of GaAs was described by many authors. Here we use the structure obtained by Jancu *et al.*[23] within the $sp^3d^5s^*$ empirical tight-binding model, which takes into account the s, p, and d orbitals for both anions and cations. As shown in Ref. 23, the inclusion of d orbitals improved considerably the description of the band structure in the vicinity of the X-high symmetry point of the Brillouin zone. The spin-orbit interactions were added to the model by including the contribution from the p valence states. The tight-binding model parameters were obtained by fitting the on-site energies and the two-center NN integrals in the Hamiltonian to the measured energies and free-electron band structure. This model reproduces correctly the density of states, effective masses, and deformation potentials, without taking into account the interactions between more distinct, e.g., next NN ions.

The (Ga,Mn)As MBE-grown layers are diluted ferromagnetic semiconductors, with the Curie temperature that depends on both the Mn magnetic ions content and the concentration of holes in the valence band. The valence-band structure of (Ga,Mn)As with a small fraction of Mn was shown to be quite similar to that of GaAs (Ref. 24), and we take most of parameters to be identical to those in GaAs. The presence of the Mn ions in the lattice results, however, in spin splittings of the conduction and valence bands, due to sp-d exchange interactions between the spins of the band electrons and localized Mn magnetic moments. These interactions are included in the tight-binding Hamiltonian using the mean-field prescription with the experimental values of the exchange integrals $N_0\beta=-1.2$ eV and $N_0\alpha=0.2$ eV (Ref. 24).

We built the SL assuming that the band offsets at the (Ga,Mn)As and GaAs interfaces are induced solely by the spin splittings in the (Ga,Mn)As bands. In structures incorporating (Al,Ga)As nonmagnetic layers large band offsets (e.g., for 30% of Al, 0.41 eV in the valence, and 0.15 eV in the conduction band) have to be taken into account. The relatively small lattice mismatch between GaAs and (Ga,Mn)As, (Ref. 25), as well as the strains resulting from it, have been ignored. All the experimentally studied (Ga,Mn)As-based SL were grown on GaAs substrate along the [001] crystallographic axis. In this case the primitive lattice vectors, which define the SL elementary cell, are: $\mathbf{a_1}=a\sqrt{3}/2[1,1,0]$; $\mathbf{a_2}=a\sqrt{3}/2[1,0,m+n]$; $\mathbf{a_3}=a\sqrt{3}/2[0,1,m+n]$. The spins in the magnetic layers are aligned along the [100] direction.[10] The SL elementary magnetic cell, which has to be considered, must contain at least two magnetic layers, i.e., it should consist of $2(n+m)$ monolayers. This, together with the used description of the constituent materials, leads to $80(m+n)\times 80(m+n)$ matrix for the SL tight-binding Hamiltonian. By numerical diagonalization of the Hamiltonian matrices, the band structure for the two SL with different relative spin configurations of the (Ga,Mn)As FM layers is obtained. The position of the Fermi level in the SL valence band is assumed to be determined by the average number of holes present in the structure—for (Ga,Mn)As/GaAs it is given by $a^3/4(p_m m)$, whereas for (Ga,Mn)As/GaAs:Be by $a^3/4(p_m m+p_n n)$. In (Ga,Mn)As/(Al,Ga)As with high Al content the holes are confined in the (Ga,Mn)As layers, due to the high potential barriers introduced in the valence band by the spacer layer. As all the studied structures contain (Ga,Mn)As layers that were not annealed, we assume the hole density in (Ga,Mn)As to be equal to $p_m=2\times 10^{20}$ cm^{-3} for the sample with 4% of Mn and $p_m=3\times 10^{20}$ cm^{-3} for the sample 6% of Mn (Ref. 26). The density of holes introduced by Be in the spacer is assumed to be $p_n=1.21\times 10^{20}$ cm^{-3} (Ref. 12). In order to calculate IEC in the spirit of Ref. 13, one has to compare the total energy of the valence electrons for two different SL, one with parallel and the other with antiparallel spin alignment in consecutive magnetic layers. These total energies were calculated by summing up all the occupied states' energies and integrating over the entire Brillouin zone. It should be noted that in this calculation the hole charge redistribution at the interfaces is not taken into account.

The strength of the interlayer coupling is given by the

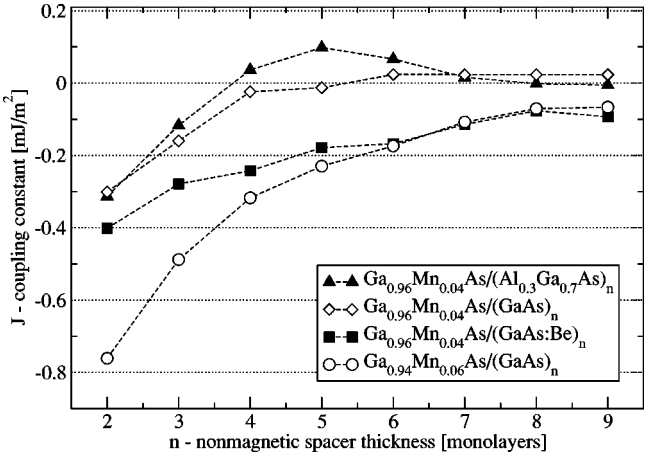

FIG. 1. The interlayer exchange coupling calculated for (Ga,Mn)As-based structures, which were studied experimentally in Refs. 6, 10, and 12.

difference ΔE between the energies of valence electrons in SL calculated for the two spin configurations, per unit surface of the layer. The preferred spin configuration in consecutive magnetic layers is given by the sign of ΔE—the negative value corresponds to FM IEC, whereas the positive sign indicates an AFM correlation. The results of the calculations are summarized in Figs. 1–3. Here, as it was in EuS-based structures, J practically does not depend on the thickness of the magnetic layer—all the presented results are calculated for $m=4$.

In Fig. 1 the calculated dependence of the interlayer coupling constant $J=\Delta E/4$ on the spacer thickness n for $Ga_{0.04}Mn_{0.06}As/GaAs$ SL is shown together with the results obtained for $Ga_{0.96}Mn_{0.04}As/GaAs$, without and with Be doping (the latter introducing p_n holes in the spacer layer) and $Ga_{0.96}Mn_{0.04}As/Ga_{0.7}Al_{0.3}As$, i.e., for the other experimentally studied (Ga,Mn)As-based structures. In qualitative agreement with the experiment, the obtained IEC for all these structures is, in principle, FM and decreases with the thickness of nonmagnetic layers. The higher the hole concentration in the SL the stronger is the IEC. For the (Ga,Mn)As/(Al,Ga)As sample, where the holes are confined in the deep wells formed by the barriers of spacer layers, the IEC is considerably suppressed and vanishes for $n>7$, as measured in Ref. 6 for the structure with $n=10$. This result does not confirm, however, that there is no IEC without holes in the spacer layer. For very thin spacers, 2-3 monolayers, a strong FM coupling, and for $n=5$ an AFM coupling were obtained (see Fig. 1).

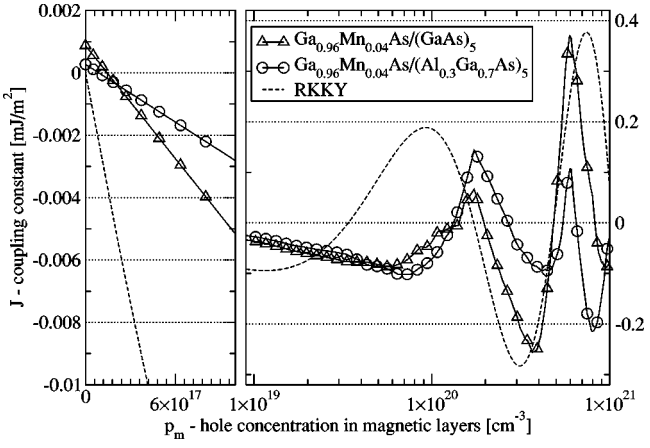

FIG. 2. The calculated dependence of interlayer coupling constant J on the hole concentration in SL consisting of alternating $m=4$ $Ga_{0.96}Mn_{0.04}As$ monolayers and $n=5$ monolayers of GaAs or $Ga_{0.7}Al_{0.3}As$. J_{RKKY} is shown for comparison.

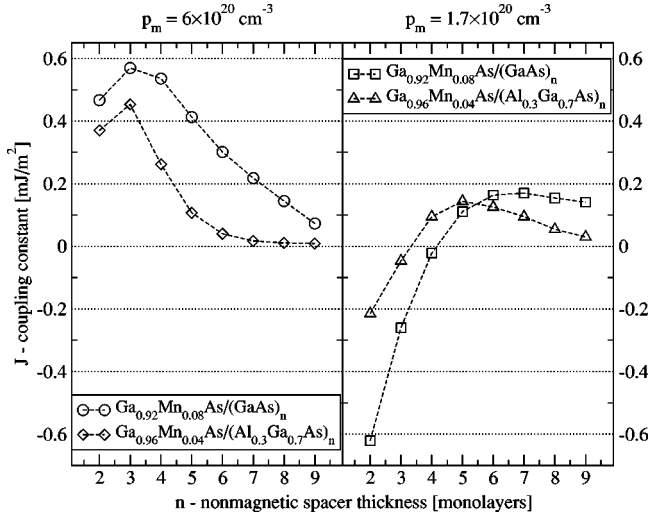

FIG. 3. The coupling constant vs spacer thickness for (Ga,Mn)As-based SL in the two regions of hole concentration, in which the model predicts an antiferromagnetic interlayer coupling.

To make these results and the role played by holes more clear, the dependence of the calculated interlayer coupling constant J on the position of the Fermi level, i.e., on the average concentration of holes in the SL valence band, was studied. As shown in Fig. 2, J has an oscillatory RKKY-like character [for comparison, IEC mediated by RKKY interaction, i.e., $J_{RKKY}\sim k_F^2F(2k_Fr)$, where k_F is the Fermi wave vector and $F(x)=(x\cos x-\sin x)/x^2$ (Ref. 27), is presented in the figure by the dashed line]. In contrast to J_{RKKY}, at the zero hole concentration limit J tends not to zero, but to a finite positive value, which corresponds to IEC mediated by valence-band electrons in a hypothetical (Ga,Mn)As/GaAs SL with completely filled valence bands. In (Ga,Mn)As/(Al,Ga)As SL, for the concentrations up to about 4×10^{20} cm^{-3}, the holes are confined in the wells—when the Fermi level reaches the value of the band offset between (Ga,Mn)As and (Al,Ga)As, the distribution of holes in the SL changes and the J values obtained for higher concentrations do not follow the previous trends. Importantly, the results presented in Fig. 2 indicate that in (Ga,Mn)As-based heterostructures also the AFM coupling between FM layers could be achieved by an appropriate engineering of the SL and a proper choice of constituent materials. The change of the IEC sign was also obtained within a kp theory with a single parabolic valence band.[21] However, on the grounds of the present results, structures particularly suitable for the observation of AFM correlations can be suggested. These seem to be SL in which the hole concentration is either increased (e.g., by appropriate annealing during the

molecular-beam epitaxy (MBE) growth of the SL) to about 6×10^{20} cm^{-3} or kept as low as $1.5-2.5\times10^{20}$ cm^{-3}. It should be noted that in the former, one can also expect high Curie temperature. The (Ga,Mn)As/(Al,Ga)As system is additionally interesting because here, due to high potential barriers in the nonmagnetic spacers, the carriers are confined in the DMS layers, and can result in strongly spin-polarized charge density. In the latter heterostructures the height of the barrier, i.e., the Al content, is very important—the results for (Ga,Mn)As/AlAs SL (for clarity not included in the figure) show that very high barriers reduce extremely the IEC in both FM and AFM regions. Finally, in Fig. 3 we show the dependencies of J on the thickness of the spacer layer n for the $Ga_{0.92}Mn_{0.08}As/GaAs$ and $Ga_{0.96}Mn_{0.04}As/Al_{0.3}Ga_{0.7}As$ SL with appropriate for AFM IEC hole concentrations. For the higher concentration the coupling is stronger for both structures, but decreases more rapidly with the spacer thickness. It should be noted that SL with the spacers as thin as 3 monolayers, for which the strongest coupling has been predicted, would be difficult to obtain, due to the strong interdiffusion in the LT MBE grown (Ga,Mn)As structures.[28] Still, for $n=5-6$, the predicted AFM IEC is of the same order of magnitude as the FM coupling observed in the (Ga,Mn)As-based SL.

In conclusion, we have studied, within a tight-binding model, the sensitivity of the band structure of (Ga,Mn)As-based SL to the spin configuration in successive DMS layers. Such effects describe correctly the AFM IEC between the FM layers in EuS/PbS and EuS/YbSe and are, up to now, the only effective mechanism capable of explaining the origin of interlayer correlations in AFM EuTe/PbTe SL. We have shown that by this mechanism also the FM interlayer coupling in (Ga,Mn)As/GaAs SL can be described. Moreover, the model points to a possibility of engineering (Ga,Mn)As-based multilayers for obtaining an AFM interlayer coupling.

The authors thank T. Story for elucidating discussions. This work was supported by the Polish Ministry of Science (PBZ-KBN-044/P03/2001) and FENIKS (EC:G5RD-CT-2001-00535), and ERATO Semiconductor Spintronics projects.

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