Interlayer Exchange Coupling in (Ga,Mn)As-based Superlattices

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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 allows also to predict (Ga,Mn)As-based structures, in which an antiferromagnetic interlayer coupling could be expected.

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Interlayer exchange coupling (IEC) – the phenomenon, which was shown to be responsible for the giant magnetoresistance effect and which led already to many applications of magnetic semiconductor thin film structures – was discovered in late 1980-s. Since the first report on correlated magnetization vectors in Fe/Cr/Fe trilayers, 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. Next, such coupling was also identified in semiconductor multilayer structures with FM, (Ga,Mn)As and EuS 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 application point of view, the most interesting was the discovery of AFM coupling between FM layers in EuS/PbS SL. 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. In (Ga,Mn)As-based FM structures, where a higher critical temperature can be achieved, unfortunately only FM IEC was observed.

To explain the spin correlations observed in the AFM EuTe/PbTe and FM EuS/PbS SL, a model was proposed 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, 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 and to an AFM coupling between successive FM EuS layers in EuS/PbS SL. 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 SQUID 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.

In the (Ga,Mn)As-based semiconductor ferromagnetic/nonmagnetic systems interlayer coupling of opposite FM sign was observed – by magnetic measurements and by neutron diffraction and polarized neutron reflectometry. 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 rock-salt crystal structure of EuS-based SL, they crystallize in 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 and confirmed by neutron reflectivity experiments in EuS/YbSe SL. Finally, (Ga,Mn)As is not a magnetic but doped magnetic semiconductor – 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 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. To explain the spin correlations between (Ga,Mn)As layers the RKKY mechanism and the models tailored for metallic systems were invoked. In this paper, in order to describe the spin-dependent band structure effects which 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. 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, to select the set of atomic orbitals for every type of involved ions and to 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 $a = 5.653 \text{ Å}$ 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, $\Delta_p = 0.34$ eV. The band structure of GaAs was described by many authors. Here we use the structure obtained by Jancu at al. within $sp^3d^5s^*$ empirical tight-binding model, which takes into account the $s$, $p$ and $d$ orbitals for both, anions and cations. As shown in Ref. 22, the inclusion of $d$-orbitals improved considerably the description of the band structure in the vicinity of 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 which 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 small fraction of Mn was shown to be quite similar to that of GaAs, 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 into 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.

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 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 [001] crystallographic axis. In this case the primitive lattice vectors, which define the SL elementary cell are: $a_1 = a\sqrt{3}/2[1, 1, 0]$; $a_2 = a\sqrt{3}/2[1, 0, m + n]$; $a_3 = a\sqrt{3}/2[0, 1, m + n]$. The spins in the magnetic layers are aligned along the [100] direction. 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. Thus, the SL elementary magnetic cell, which has to be considered, must contain at least two magnetic layers, i.e., it should consists of $2(n + m)$ monolayers. This together with the used description of the constituent materials leads to $80(m + n) \times 80(n + m)$ matrix for the SL tight-binding Hamiltonian. After the numerical diagonalization of the two Hamiltonian matrices, which correspond to the two different relative spin configurations of the (Ga,Mn)As FM layers, the SL occupied states’ energies were summed up to the Fermi energy and integrated over the entire Brillouin zone. 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...
and with Be-doping (the latter introducing the spacer layer) and Ga
the results obtained for Ga
n
for Ga
siderably suppressed and vanishes for
formed by the barriers of spacer layers, the IEC is con-
valence band by the spacer layer. As all the studied
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. For very thin spacers, 2-3 monolayers,
AFM correlations can be suggested. These seem
appropriate engineering of the SL and a proper choice of
in (Ga,Mn)As-based heterostructures also the AFM cou-
the previous trends. Importantly, as suggested before
obtained
J
F
RKKY
for the sample 6% of Mn.
The density of holes introduced by Be in the spacer is
assumed to be
p
n
2.1 × 10
20
cm
−3
12

The strength of the interlayer magnetic coupling is
given by
(p
m
− m, n), whereas for (Ga,Mn)As/GaAs:Be by
(m, n)

2

J
F
RKKY
is shown for comparison.

In Fig. 1 the calculated dependence of the interlayer coupling constant
J = \Delta E/4 on the spacer thickness
n for Ga
0.94
0.06
As/GaAs SL is shown together with the results obtained for Ga
0.04
0.04
As/GaAs, without and with Be-doping (the latter introducing
p
h
holes in the spacer layer) and Ga
0.04
0.7
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 nonmag-
netic layers. The higher the hole concentration in the SL the stronger is the ICE. 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 con-
siderably suppressed and vanishes for
n > 7, as measured in Ref. 11 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 was obtained (see Fig. 1).

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^2 F(2k_F r),
where
k_F
is the Fermi wave vector and
F(x) = (x \cos x - \sin x)/x^2
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 ob-
tained
J values for higher concentrations do not follow the previous trends. Importantly, as suggested before in Ref. 11, the presented in Fig. 2 results 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. On the grounds of the presented 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 in-
creased (e.g., by appropriate annealing during the MBE growth of the SL) to about
6 \times 10^{20}cm^{-3}
in the SL, where the holes are confined in the deep wells formed by the barriers of spacer layers, the IEC is con-
siderably suppressed and vanishes for
n > 7, as measured in Ref. 11 for the structure with
n = 10. This result does

FIG. 1: The interlayer exchange coupling calculated for
(Ga,Mn)As-based structures, which were studied experimentally in Refs. 4, 11 and 12.

FIG. 2: The calculated dependence of interlayer coupling constant
J on the hole concentration in SL consisting of alternating
m = 4 Ga
0.06
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.

because here, due to high potential barriers in the nonmagnetic spacers, the carriers are confined in the DMS layers, what 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$_{0.98}$Mn$_{0.02}$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 it 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 to explain 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.

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1. S. S. P. Parkin, Phys. Rev. Lett. 67, 3598 (1991).
2. G. A. Prinz, Science 282, 1660 (1998).
3. P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky and H. Sower, Phys. Rev. Lett. 57, 2442 (1986).
4. P. Bruno, Phys. Rev. B 52, 411 (1995).
5. V. Nunez et al., J. Magn. Magn. Mat. 140-144, 633 (1995); T.M. Giebultowicz et al., ibidem p. 635.
6. N. Akiba, F. Matsukura, A. Shen, Y. Ohno, H. Ohno, A. Oiwa, S. Katsumoto and Y. Iye, Appl. Phys. Lett. 73, 2122 (1998).
7. H. Kępa et al., Europhys. Lett. 56, 54 (2001).
8. A. Stachow-Wojcik et al., Phys. Rev. B 60, 15220 (1999).
9. D. Chiba, N. Akiba, Y. Ohno and H. Ohno, Appl. Phys. Lett. 77, 1873 (2000).
10. H. Kępa, J. Kutner-Piaśzek, A. Twardowski, C. F. Majkrzak, J. Sadowski, T. Story and T.M. Giebultowicz, Phys. Rev. B 64, 121302(R) (2001).
11. W. Szauskiewicz et al., Acta Phys. Pol. A 100, 335 (2001).
12. S.J. Chung, S. Lee, I.W. Park, X. Liu and J.K. Furdyna, J. Appl. Phys. 95, 7402 (2004).
13. J. Blinowski and P. Kacman, Phys. Rev. B 64, 045302 (2001).
14. H. Kępa et al., Phys. Rev. B 68, 024419 (2003).
15. C.J.P. Smits et al., Phys. Rev. B 69, 224410 (2004).
16. M. Chernyshova et al., Acta Phys. Polon. A 105, 599 (2004).
17. H. Kępa, P. Sankowski, P. Kacman, C.F. Majkrzak and T.M. Giebultowicz, Proc. XXVII ICPH, Flagstaff, Arizona 2004 (in print).
18. P. Sankowski and P. Kacman, Acta Phys. Pol. A 103, 621 (2003).
19. H. Kępa, P. Sankowski, P. Kacman, A. Yu. Sipatov, C.F. Majkrzak and T.M. Giebultowicz, J. Magn. Magn. Mat. 323, 272-276 (2004).
20. T. Dietl, H. Ohno, F. Matsukura, J. Cibert and D. Ferrand, Science 287, 1019 (2000).
21. T. Jungwirth, W.A. Atkinson, B.H. Lee and A.H. MacDonald, Phys. Rev. B 59, 9818 (1999).
22. M. A. Boselli, I. C. da Cunha Lima and A. Ghazali Phys. Rev. B 68, 085319 (2003).
23. J-M Jancu, R. Scholz, F. Beltram and F. Bassani, Phys. Rev. B 57, 6493 (1998).
24. J. Okabayashi et al., Physica E 10, 192 (2001).
25. I. Kuryliszyn-Kudelska, J. Domagala, T. Wojtowicz, X. Liu, E. Lusakowska, W. Dobrowolski and J.K. Furdyna, J. Appl. Phys. 95, 603 (2004).
26. V. Osinniy et al., http://arXiv.org/cond-mat/0409659.
27. Y. Yafet, Phys. Rev. B 36, 3948 (1987).
28. A. Mikkelsen et al., Appl. Phys. Lett. 85, 4660 (2004).