Interlayer Exchange Coupling Mediated by Valence Band Electrons

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The interlayer exchange coupling mediated by valence band electrons in all-semiconductor IV-VI magnetic/nonmagnetic superlattices is studied theoretically. A 3D tight-binding model, accounting for the band and magnetic structure of the constituent superlattice components is used to calculate the spin-dependent part of the total electronic energy. The antiferromagnetic coupling between ferromagnetic layers in EuS/PbS superlattices is obtained, in agreement with the experimental evidences. The results obtained for the coupling between antiferromagnetic layers in EuTe/PbTe superlattices are also presented.

I. INTRODUCTION

Interlayer exchange coupling (IEC) was discovered in late 80-ties in Fe/Cr/Fe trilayer. Since then it has been observed in a variety of multilayer structures composed of alternating magnetic and nonmagnetic layers. These studies concentrated on the coupling between ferromagnetic, metallic layers, although separated by both metallic and insulating spacers. Thus, the ferromagnetic character of the magnetic layers and the fact that in these structures the Fermi level was situated in the region of high density of electronic states inherited to the theoretical models, which were designed to explain the origins of the IEC phenomena (see and the references therein). Surprisingly enough, the IEC was also discovered in all-semiconductor superlattices (SLs). Moreover, the semiconductor SLs in which it was first observed were the MnTe/CdTe, MnTe/ZnTe, and EuTe/PbTe all with antiferromagnetic layers. Recently, such coupling was also identified in semiconductor multilayer structures with ferromagnetic EuS and Ga(Mn)As layers. While the IEC in the trilayer Ga(Mn)As/Ga(Al)As/Ga(Mn)As with the high concentration of free carriers can be, at least qualitatively, explained in terms of the models tailored for metallic systems, the other semiconductor structures exhibiting IEC call for a different approach.

Several attempts to explain IEC in all-semiconductor structures have already been reported in the literature. Two models, in which the interlayer coupling is mediated by carriers localized on shallow impurities in the spacer region, were proposed for II-VI SLs. These models do not apply to IV-VI structures, with the PbTe and PbS spacers, since in lead chalcogenides localized shallow impurity states were never detected. For these SLs, mechanisms of interlayer spin-spin interactions mediated by valence-band electrons were suggested. The calculations of the difference between total electronic energies obtained for two different (B1 and B2 in Fig. 1) spin configurations of the SL, performed within a frame of a very simple 1D tight-binding model, put first in evidence the significant role of the valence band electrons in IEC in all-semiconductor magnetic/nonmagnetic layer structures. This role was further demonstrated for EuTe/PbTe/EuTe trilayers by Wilczyński and Świrkowski in, where a 3D tight-binding model, still oversimplifying the band structure, was used. A different approach to the magnetic interlayer interactions mediated by valence electrons have been chosen by Dugaev et al. These authors studied the Blombergen-Rowland mechanism within the effective mass approximation and obtained a ferromagnetic coupling between two magnetic impurities situated at the opposite interfaces of a narrow-gap IV-VI semiconductor spacer. As the experimentally observed IEC in EuS/PbS SLs is antiferromagnetic, this means that the Blombergen-Rowland interactions are not dominating IEC in these SLs. Still, because of the low concentration of free carriers and the absence of shallow impurity states in PbS, the IEC in these SLs is most likely mediated by valence electrons. In this situation, the total energy calculations, which do not focus on a particular interaction mechanism but account globally for the spin-dependent structure of valence bands, seem to constitute the most appropriate approach. The calculations of this type, reported in and, were clearly oversimplified and performed for a different spin structure than that of EuS/PbS (001) SL.

In this paper we present the results of refined 3D total energy calculations, which take into account the crystal and the band structures of the SLs’ component materials. The tight-binding model with its assumptions and the results for three different spin structures, corresponding to all experimentally studied IV-VI semiconductor magnetic/nonmagnetic SLs, are presented in Section II. In Section III the magnetic and band structures of the constituent materials are described and the SL geometry is specified. The comparison with experimental data and the conclusions are presented in the last Section.
II. CONSTITUENT MATERIALS AND THE SUPERLATTICES GEOMETRY

All the components of the EuX/PbX SLs, where X=Te or S, crystallize in the rock-salt structure. Bulk PbS and PbTe are narrow gap nonmagnetic semiconductors with very similar band structures. They both have a direct energy gap between the p-anion valence band and the p-conduction band at the point L of the BZ. It is well known that in both these lead chalcogenides the spin-orbit terms are important for the detailed description of the energy bands in the vicinity of the L point. One notices, however, that the spin-orbit corrections affect predominantly the conduction bands originating from the states of the heavy Pb atoms.

Bulk EuS is a classical Heisenberg ferromagnet with the Curie temperature 16.6 K. Bulk EuTe exhibits type II AFM structure with the Néel temperature 9.6 K. In EuTe the spins of Eu ions are ferromagnetically ordered in (111)-type planes, which in turn are coupled antiferromagnetically to one another. All the Eu chalcogenides are semi-insulating, large gap semiconductors. The results of the nonrelativistic APW calculations of the EuS spin-polarized band structure show the narrow f↑ bands situated in the energy gap between the valence band, formed essentially of anion p states, and the conduction band, built mostly of cation d states. The valence band maximum is situated at the center of the Brillouin zone (BZ) and the conduction band minimum at the point X. The spin splitting of the valence band results predominantly from the spin dependent mixing of p anion and f cation states, whereas that of the conduction band is mostly due to f-d and s-d on-site direct exchange. Much less is known about the EuTe band structure. The optical experiments performed at T=300 K indicate that in the paramagnetic phase of EuTe the f-d gap is somewhat larger than in EuS, in agreement with the general trends visible in the experimental data and the results of APW calculations for other europium chalcogenides. These trends seem not to be followed in the recent calculations of EuTe band structure, focused predominantly on the conduction bands.

Two types of EuS/PbS SLs were experimentally studied, one grown on KCl substrate along [001] and the other on BaF₂ along the [111] crystallographic axis. The measurements show that in both cases the ferromagnetically ordered Eu spins within each magnetic layer lie in the planes perpendicular to the growth axis. In the (001) structures each atomic monolayer consists of both anions and cations, with the monolayers a/2 apart, where a is the cubic lattice constant. The schematic view of two such monolayers is presented in Fig. 3. The distances between the cation and its four in-plane nearest neighbors (anions) and four in-plane next nearest neighbors (cations) are also shown in the figure. The spin structure of the magnetic (001) SL, which corresponds to the observed antiferromagnetic interlayer coupling is shown schematically in Fig. A2.

In the case of (111) EuS/PbS SLs IEC has not been yet observed. These SLs have the same crystallographic structure as the experimentally studied EuTe/PbTe SLs grown along the [111] axis. With this growth direction, the subsequent (111) atomic monolayers are built either of anions or of cations, in alternation. The distance between cation and anion monolayers is $a\sqrt{3}/6$. A schematic view of three successive cation layers is presented in Fig. B - the analogous anion sublattice is shifted by $a\sqrt{3}/2$ along the [111] direction and is not shown in the figure for the clarity reasons.

In EuTe/PbTe SLs the neutronographic measurements show that the AFM Type II structure is preserved in each EuTe layer, but the FM spin sheets form exclusively on the (111) planes parallel to the layers. This is in contrast to bulk EuTe, where there are four symmetry-equivalent Type II AFM arrangements in which the ferromagnetic spin sheets form on the {111}, {1T1}, {1T1}, or {11T} plane families. Moreover, for the nonmagnetic spacers thin enough, the satellites observed in neutronographic spectra clearly indicate the existence of some long range order proving that the spins in consecutive magnetic layers are not randomly oriented, but tend to align along the same direction in a correlated way. Although for the antiferromagnetic layers the notions of AFM and FM IEC are not applicable, two types of co-linear correlated spin orientations in successive layers are still possible: identical (in-phase) and reversed (out-of-phase), as shown in Fig. B. Both types of IEC were observed in the experiment.

All the (EuX)ₘ/(PbX)ₙ SLs (where m and n denote the number of EuX and PbX layers, respectively) have
To discuss the spin coupling between the magnetic layers one has to consider a SL magnetic elementary cell containing at least two such layers. In SLs grown along [001] crystallographic direction the situation is simple, as the stacking sequence is ABAB-type. Such stacking does not enlarge the size of the magnetic elementary cell, whatever \( m + n \) value. In contrast, the stacking sequence ABCABC-type (compare Fig. 3) for both anions and cations, in SLs grown along [111] axis does enlarge the elementary cell when \( m + n \) is not a multiple of 3. Thus, to limit the size of elementary cells, we consider only the (111) SLs with \((n + m)/3 = \text{integer}\).

In the case of (001) SLs the primitive lattice vectors, which define our elementary cells are: \( \mathbf{a}_1 = a/2[1, -1, 0]; \mathbf{a}_2 = a/2[1, 1, 0]; \mathbf{a}_3 = a[0, 0, m + n] \). For (111) SLs, when the z-axis is chosen along the growth direction, we have \( \mathbf{a}_1 = a/4[\sqrt{2}, -\sqrt{6}, 0]; \mathbf{a}_2 = a/4[\sqrt{2}, \sqrt{6}, 0]; \mathbf{a}_3 = (2a/\sqrt{3})[0, 0, m + n] \) (the plane views of these cells are sketched in Fig. 2 and Fig. 3). For both types of SL, our magnetic elementary cells contain, therefore, \( 2(m + n) \) anions, \( 2m \) magnetic cations and \( 2n \) nonmagnetic cations.

In order to determine IEC in the above structures we compare the total valence-electron energies in two magnetic SLs: with the in-phase and out-of-phase spin ordering. For the \( n \) and \( m \) values typical for the experimentally studied SLs the elementary cells contain several tens of atoms. In view of this complexity, we decide to use the simplest calculation scheme still leading to fairly realistic band structure, namely, an empirical tight-binding method. Even though the one-electron methods are not designed for the total-energy calculations, the small spin-dependent changes in the total energy should be described adequately within this approach.

### III. TIGHT-BINDING MODEL

To construct the empirical tight-binding Hamiltonian matrix one has to select the set of atomic orbitals for every type of involved ions and to specify the range of the ion-ion interactions. This selection is always a compromise between the best description of the band structure and the minimization of the Hamiltonian matrix dimensions and of the number of parameters used. 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 known band structures of the bulk constituent magnetic and nonmagnetic materials, respectively. This criterion determines in principle the selection of the ionic orbitals and gives the values of the parameters, all but those characterizing the interaction between magnetic and nonmagnetic cations.

To calculate the band structure of the lead chalcogenides (PbS and PbTe) we took into account s and p orbitals for both anions and cations, which lead to 8x8 Hamiltonian matrix. We allowed for the s–s, s–p and p–p anion–cation nearest-neighbor (NN) interactions and the anion–anion and cation–cation p–p next-nearest neighbor (NNN) interactions. It turned out that the band structure can be reproduced much better when we include also, by second-order perturbation, the interactions of p-orbitals with the three NN d-orbitals belonging to the \( F_2 \) representation. The values of the parameters describing all these interactions and the values of the on-site orbital energies were determined by a \( \chi^2 \) minimization procedure, in which the band structure was fitted to the energies in the high symmetry points of the BZ, taken from\(^{16,17} \). The obtained energy bands for PbS and PbTe along the symmetry axes of the BZ, are presented in Fig. 4.
In the other limit, for europium chalcogenides (EuS and EuTe), to describe the cations we take explicitly the one s- and five d-orbitals, whereas the anions are described as before by s- and p-orbitals. The NN interaction involving the anion p-orbitals and cation s- and d-orbitals as well as the NNN cation-cation d–d and anion-anion p–p interaction were included in the 10x10 Hamiltonian. The s-anion–s-cation, s-anion–d-cation interactions, turned out to be less important and were neglected. Instead, we included, again by second order perturbation, the hybridization of anion p(↑)-orbitals with the cation f(↑)-orbitals - this was necessary for reproducing the spin splittings of the valence bands in the ferromagnetic EuS (we neglected the hybridization with the energetically distant f(↓) band). To reproduce the spin splittings in the EuS conduction bands the on-site exchange constants \( J_s \) and \( J_d \) had to be introduced. The band structure of EuS, presented in Fig. 5 was obtained with the parameters fitted to the results of the APW spin–polarized calculations reported in \(^{18}\). The presented in Fig. 5 band structure of EuTe was obtained with the parameter values extrapolated from the values for EuS and EuSe by exploiting the chemical trends in europium chalcogenides. The elementary cell of the antiferromagnetic EuTe has a twice larger volume and completely different shape than the one of the ferromagnetic EuS – to facilitate the comparison with EuS, we present the band structure of EuTe in the paramagnetic phase.

In principle, the SL tight-binding Hamiltonian is therefore a \((40m + 28n)\times(40m + 28n)\) matrix, which has to be completed by the nonmagnetic cation–magnetic cation interactions. The constants describing these interaction can not be inferred from the \( m = 0 \) and \( n = 0 \) limits, they were estimated from Harrison’s formula for interatomic matrix elements. The matrix was then perturbationally reduced to the \((20m + 16n)\times(20m + 16n)\) matrix.

To determine the small difference between the large total energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately. Instead, the difference between the two energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately. Instead, the difference between the two energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately. Instead, the difference between the two energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately. Instead, the difference between the two energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately. Instead, the difference between the two energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately. Instead, the difference between the two energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately. Instead, the difference between the two energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately. Instead, the difference between the two energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately. Instead, the difference between the two energies of the valence electrons in the two, in-phase and out-of-phase, spin configurations, we did not calculate these energies separately.
in the case of the hexagonal BZ of the (111) SLs, were then used in the triple Simpson procedure for integrating over the entire BZ. The calculations were performed for (EuS)$_m$/(PbS)$_n$ SLs grown in both [001] and [111] directions and for (EuTe)$_m$/(PbTe)$_n$ SLs grown along the [111] axis.

![Model band structures of ferromagnetic EuS and paramagnetic EuTe](image)

**FIG. 5:** Model band structures of ferromagnetic EuS (solid lines represent the spin-down bands; dotted lines – spin-up bands) and paramagnetic EuTe

We denote by $\Delta E$ the absolute value of the energy difference between the in-phase and out-of-phase spin configuration per unit surface of the layer. $\Delta E$ can be regarded as a measure of the strength of the interlayer spin coupling in the SLs – for ferromagnetic structures it can be expressed in terms of the constant $J_{1,2,4}$ commonly used to characterize the IEC in metallic magnetic/nonmagnetic trilayers by the relation: $\Delta E = 4|J_{1}|$ (here the factor 4, instead of 2, accounts for the fact, that in SL each magnetic layer is coupled to two neighboring layers).

The sign of the calculated energy difference determines the spin configuration in consecutive magnetic layers. In the ferromagnetic, EuS-based, SLs the out-of-phase spin configuration in consecutive magnetic layers is energetically preferred, so that IEC in these structures has an ANTIFERROMAGNETIC character, in agreement with the experiment. For the antiferromagnetic, EuTe-based SLs the situation is more complicated: for odd number $m$ of spin planes in the magnetic layer the out-of-phase configuration has the lower energy, whereas for even $m$ it is the in-phase configuration, which is energetically favored. Thus, one can notice that in all studied SLs the valence electron mediated IEC prefers the spin configuration with the opposite directions of spins at the two interfaces bordering the spacer.

Many various $m$ and $n$ values were selected to study the range of the interlayer coupling and the IEC dependence on the thickness of the magnetic layer. It turned out, that in all SLs for fixed spacer thickness $n$ the strength of IEC is almost independent on the magnetic layer thickness $m$. This seems to prove that in the considered here SLs, composed of two semiconductors with very different energy gaps, the valence electron mediated IEC is essentially a surface effect.

The dependence of the strength of the interlayer coupling on the spacer thickness $n$ for all three studied types of SLs is presented in Fig. 6. We recall that for the [111] growth direction, we calculated IEC only for $(n + m)/3 = integer$, so that for these SLs in the figure the points for different $n$ values do not correspond to the same $m$. As can be seen in Fig. 6, the strength of the coupling in all three cases decreases with the spacer thickness approximately exponentially. The strongest and the least rapidly decreasing IEC was obtained in the case of the FM EuS/PbS (001) SLs. The comparison of the results obtained with the same set of model parameters for the two different types of EuS/PbS SLs, (i.e., the
The IEC was observed in ferromagnetic EuS/PbS (001) SLs by magnetic and neutron diffraction and reflectivity methods. For the samples with the thin enough spacers, i.e., 4.5 Å (1.5 monolayers, probably a mixture of $n = 2$ and $n = 1$) and 10 Å (ca $n = 3$), the antiferromagnetic interlayer coupling was observed in the magnetic moment measurements; the magnetic methods did not reveal any IEC in the samples with larger spacer thicknesses. In the neutronographic experiments the AFM IEC was confirmed in the above two samples, but it was also observed in the sample with 23 Å PbS layers. Further measurements in the external magnetic fields, parallel to the layers, allowed to estimate the experimental strength of the coupling constant $J_1$, from the magnetic field $B$ erasing the AFM neutron reflectivity peak. This was possible for the two samples with the thinner spacers, but not for the sample with the thicker spacer. For the latter, the field-induced changes in the neutronographic spectra were irreversible, what suggests that in this case the IEC was weaker than the magnetic anisotropies. The estimated experimental values of $J_1$ are: 0.063, 0.031, and 0.019 (in mJ/m$^2$), for $n=1$, 2 and 3, respectively. The corresponding theoretical values obtained from our model are: 0.77, 0.33, 0.18 mJ/m$^2$. Thus, one can conclude, that the model of valence electron mediated IEC describes properly the sign and the rate of the decrease of the coupling with the spacer thickness, but overestimates the strength of the coupling. The fact that the theoretical results obtained for crystallographically perfect SLs lead to exchange constants order of magnitude larger than those observed for the real multilayer structures, is probably due to the interface diffusion, which in the case of metallic structures was shown to reduce significantly the strength of the IEC.

### IV. COMPARISON WITH THE EXPERIMENTAL DATA

#### A. Ferromagnetic EuS/PbS superlattices

The IEC was observed in ferromagnetic EuS/PbS (001) SLs by magnetic and neutron diffraction and reflectivity methods. For the samples with the thin enough spacers, i.e., 4.5 Å (1.5 monolayers, probably a mixture of $n = 2$ and $n = 1$) and 10 Å (ca $n = 3$), the antiferromagnetic interlayer coupling was observed in the magnetic moment measurements; the magnetic methods did not reveal any IEC in the samples with larger spacer thicknesses. In the neutronographic experiments the AFM IEC was confirmed in the above two samples, but it was also observed in the sample with 23 Å PbS layers. Further measurements in the external magnetic fields, parallel to the layers, allowed to estimate the experimental strength of the coupling constant $J_1$, from the magnetic field $B$ erasing the AFM neutron reflectivity peak. This was possible for the two samples with the thinner spacers, but not for the sample with the thicker spacer. For the latter, the field-induced changes in the neutronographic spectra were irreversible, what suggests that in this case the IEC was weaker than the magnetic anisotropies. The estimated experimental values of $J_1$ are: 0.063, 0.031, and 0.019 (in mJ/m$^2$), for $n=1$, 2 and 3, respectively. The corresponding theoretical values obtained from our model are: 0.77, 0.33, 0.18 mJ/m$^2$. Thus, one can conclude, that the model of valence electron mediated IEC describes properly the sign and the rate of the decrease of the coupling with the spacer thickness, but overestimates the strength of the coupling. The fact that the theoretical results obtained for crystallographically perfect SLs lead to exchange constants order of magnitude larger than those observed for the real multilayer structures, is probably due to the interface diffusion, which in the case of metallic structures was shown to reduce significantly the strength of the IEC.

### B. Antiferromagnetic EuTe/PbTe superlattices

Unfortunately, for the AFM type of SLs there are no experiments, which provide direct information about the strength of the coupling. The evidences of the existence of the coupling between the AFM EuTe layers come from the satellite structure of the neutron diffraction spectra, seen in a variety of EuTe/PbTe SLs consisting of several hundreds of periods. The detailed analysis of the shapes of the satellite lines in the neutronographic spectra indicates that in these SLs the EuTe layers are not entirely coupled, but only partially correlated, the less the thicker are the PbTe spacer layers of the SL. Under the strong assumption that the structures are morphologically perfect, with the same $m$ and $n$ values throughout the entire SL, this degree of correlation can be quantitatively determined. Under the same assumption, the analysis of the satellites positions allows to distinguish which spin configuration, the in-phase or out-of-phase, is dominating.

The observed spectra for the SLs with nominally even $m$ and even $n$ reveal the preference for the in-phase spin configurations, whereas those for the SLs with odd $m$ and even $n$ exhibit the preference for the out-of-phase configuration, both in agreement with the predictions of
the present model. For the case of even \( m \) and odd \( n \) there are no available data. Finally, for the samples with \( m \) and \( n \) both odd, the neutron diffraction spectra seem to indicate that the in-phase configuration is preferred, contrary to the theoretical predictions. However, the in-phase spin configuration for SLs with odd number of spin planes in each antiferromagnetic layer should exhibit ferromagnetic properties, i.e., lead to a significant net magnetic moment of ferrimagnetic domains. No such magnetic moments were detected in these samples. These somewhat confusing results seem to indicate that both chemical and magnetic structures of the studied SLs are not perfect enough. New technological and experimental efforts to observe the IEC in EuTe/PbTe SLs with smaller number of SL periods, i.e., in SLs with better controlled \( m \) and \( n \) values, are undertaken.

In conclusion, we have shown, within a 3D tight binding model, that the valence electron mediated interlayer exchange coupling explains the AFM coupling between the FM layers observed in EuS/PbS (001) SLs with narrow PbS spacers. The strength of the calculated depends strongly on the lattice geometry and decreases approximately exponentially with the spacer thickness \( n \). For a given type of SL it is almost independent on the number \( m \) of the spin planes within each magnetic layer. These features distinguish the considered mechanism from another mechanism of AFM coupling between the FM layers, namely the dipolar coupling possible in multilayer structures with tiny magnetic domains.

The valence electron mediated interlayer coupling is, up to now, the only effective mechanism capable to explain the origin of the interlayer correlations observed in the antiferromagnetic EuTe/PbTe SLs with no localized impurity states. The current, not complete understanding of the experimental data for AFM SLs, does not allow, however, to draw definite conclusions about the comparison between the details of the experimental and theoretical results.

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