(Ga,Mn)As based superlattices and the search for antiferromagnetic interlayer coupling

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Antiferromagnetic interlayer coupling in dilute magnetic semiconductor superlattices could result in the realisation of large magnetoresistance effects analogous to the giant magnetoresistance seen in metallic multilayer structures. In this paper we use a mean-field theory of carrier induced ferromagnetism to explore the multidimensional parameter space available in (Ga,Mn)As based superlattice systems. Based on these investigations we examine the feasibility of creating a superlattice that exhibits antiferromagnetic coupling and suggest potentially viable recipes.

PACS numbers: 73.61.Ey, 75.50.Pp, 75.70.Cn

I. INTRODUCTION

The exciting new prospect of spin based electronics, known as spintronics, was initiated in 1988 with the discovery of giant magnetoresistance (GMR) in metallic multilayer structures. These structures consist of interposed ferromagnetic (FM) and non-FM layers. When the magnetisation of adjacent FM layers is aligned in antiparallel directions, enhanced spin scattering of carriers causes an increased electrical resistance through the layers, while when they are parallel the resistance is lower. Although typical GMR devices today consist of a trilayer structure with a pinned magnetic layer and one in which the magnetisation is free to rotate, another method of implementation is with a superlattice structure where adjacent layers have an antiparallel magnetisation unless an external field is applied to align them.

In multilayer structures containing ferromagnetic layers, in addition to the ferromagnetic order within the layers, there can also exist magnetic exchange between the layers. The mechanism which causes the magnetic order between the layers is known as interlayer exchange coupling (IEC), and has been shown in metallic systems to be due to the spin polarisation of conduction carriers.

Because the IEC energy considers the spin dependent changes in total energy, it thus determines which magnetic alignment of adjacent layers is energetically favourable. Although complicated helical arrangements can exist typically the interlayer exchange coupling will either be FM, where there is a parallel alignment of magnetisation, or antiferromagnetic (AFM) where there is an antiparallel alignment. Therefore, in such a system, achieving AFM interlayer coupling is of high importance for technological applications.

In addition to existing in metal systems, IEC is a generic property of magnetic multilayers, and AFM IEC has even been demonstrated in non-metallic FM semiconductor systems based on all-semiconductor EuS/PbS superlattices. AFM IEC in dilute magnetic semiconductor (DMS) based superlattices was theoretically predicted in 1999 using a k · p kinetic-exchange model for carrier mediated ferromagnetism. This approach considers delocalised charge and adds extra modulation induced by spin-polarised effects. A large magnetoresistance (MR) was predicted due to the large difference in miniband dispersion for the cases of ferromagnetically and antiferromagnetically aligned layers. Recently, IEC has been further explored using a tight-binding model. This complementary microscopic approach, although not self-consistent, takes into account atomic orbitals for all the constituent atoms, leading to more accurate descriptions of the band structure. Despite the different approaches used, both methods provide qualitatively similar results for the IEC which shows oscillatory Ruderman-Kittel-Kasuya-Yosida (RKKY)-like behaviour.

Although IEC has been shown to exist in DMS systems based on (Ga,Mn)As/(Al,Ga)As trilayers, there have been no reports of AFM interlayer coupling. Experimental work into (Ga,Mn)As based multilayer and superlattice structures has only succeeded in demonstrating FM IEC. In order to test the prediction of a phenomenon analogous to GMR in metals in DMS materials, with a potentially much greater MR ratio, it is essential that AFM interlayer coupling is obtained.

The aim of this study is to provide a comprehensive description of the multidimensional parameter space available in these DMS superlattice systems, in order to identify optimal parameters for realising an antiferromagnetically coupled system. Because the interlayer coupling is mediated by carriers, a k · p approach is more practical for exploring a wide range of parameter values. The limitation of this approach is that a single parabolic band approximation is used, sacrificing full quantitative accuracy for qualitative descriptions of a wide range of systems. Subtleties of the band-structure and spin-orbit effects are neglected. However, qualitative agreement with the data published in Ref. at least partially justifies this approach.

The organisation of this paper is as follows: first the details of the theoretical modelling of a DMS based superlattice system and the numerics of the self-consistent mean field calculations will be shown. Next, the results, which will primarily consider (Ga,Mn)As based su-
perlattice systems with either GaAs or (Al,Ga)As non-magnetic spacer layers, will be presented. Finally, in the discussion, suggestions for recipes for superlattice systems in which antiferromagnetic interlayer coupling may occur will be given.

II. THEORETICAL MODELLING

Our calculations are based on the Zener kinetic-exchange model\(^2\) description of magnetic interactions in Mn-doped III-V semiconductor structures. Microscopically, the kinetic-exchange between the local Mn moments and itinerant hole spins originates from the \(p-d\) orbital hybridisation.\(^3\) This model provides a good description of ferromagnetism in bulk (Ga,Mn)As when the detailed structure of the valence band is taken into account.

An intuitive picture of the IEC in (III,Mn)V/III-V multilayer structures can be obtained by the perturbative mapping of the kinetic-exchange model onto an effective interaction between local moments, following the RKKY-like oscillatory effects in \((\text{Ga,Mn})\text{As}\) was generalized in Ref.\(^7\). The Zener kinetic-exchange model provides a good description of ferromagnetism in bulk (Ga,Mn)As when the one-dimensional time-independent spin-dependent Schrödinger equation:

\[
\frac{p^2}{2m^*} + V_\sigma(z) \Psi_{k,n,\sigma}(z) = E_{k,n,\sigma} \Psi_{k,n,\sigma}(z),
\]

which we shall rewrite as

\[
\frac{d^2 \Psi_{k,n,\sigma}(z)}{dz^2} = \frac{2m^*}{\hbar^2} \left( V_\sigma(z) - E_{k,n,\sigma} \right) \Psi_{k,n,\sigma}(z),
\]

where \(k\) is the wavevector, \(n\) is the subband index, and \(\sigma\) is the spin index and \(f = \frac{2m^*}{\hbar^2} (V_\sigma(z) - E)\). The Bloch function

\[
\Psi_{k,n,\sigma}(z) = u_{k,n,\sigma}(z)e^{ikz},
\]

gives the solutions of the Schrödinger equation for a periodic potential.

For this system, the explicit form of the Hamiltonian for the spin-dependent potential \(V_\sigma(z)\) is given by

\[
V_\sigma(z) = V_H + V_{xc,\sigma} + V_b - \frac{\sigma}{2} \left[ g^* \mu_B B + h_{pd}(z) \right],
\]

where \(V_H\) is the Hartree (electrostatic) potential, given by the Poisson equation, \(V_{xc,\sigma}\) is the spin-dependent exchange-correlation potential given by the LSDA equation, \(V_b\) is the band-offset, \(g^*\) is the free-carrier g-factor and \(h_{pd}\) is the mean-field kinetic-exchange interaction.\(^2\)

The IEC energy, \(E_c\), is defined as the difference in energy between the FM and AFM states per superlattice period.

Let us suppose that our one-dimensional lattice has a period \(d_{n+m}\) and consider now the solution only at \(N\) evenly distributed discrete points on the z-axis with a separation \(h\). The wavefunction at each point is denoted as \(\Psi(z)\). By Taylor’s theorem, the second-order approximations for \(\Psi(z + h)\) and \(\Psi(z - h)\) are

\[
\Psi(z + h) = \Psi(z) + h \Psi'(z) + \frac{h^2}{2} \Psi''(z); \quad (5)
\]

\[
\Psi(z - h) = \Psi(z) - h \Psi'(z) + \frac{h^2}{2} \Psi''(z). \quad (6)
\]

Taking the sum of Eqs.\(^5\) and\(^6\) we obtain

\[
h^2 \Psi''(z) = \Psi(z + h) + \Psi(z - h) - 2\Psi(z). \quad (7)
\]

Substituting the Schrödinger equation from Eq.\(^2\) into Eq.\(^7\) and rearranging gives the wavefunction at a given point as a linear combination of the wavefunctions at the two previous points:

\[
\Psi(z + h) = (h^2 f + 2) \Psi(z) - \Psi(z - h). \quad (8)
\]

This linear transformation can be represented as a transfer matrix, \(M_n\), such that

\[
M_n = \begin{pmatrix}
1 & h^2 f + 2 \\
1 & 1
\end{pmatrix}
\]
\[ M_n \begin{pmatrix} \Psi_n \\ \Psi_{n-1} \end{pmatrix} = \begin{pmatrix} m_{n,11} & m_{n,12} \\ m_{n,21} & m_{n,22} \end{pmatrix} \begin{pmatrix} \Psi_n \\ \Psi_{n-1} \end{pmatrix} = \begin{pmatrix} \Psi_{n+1} \\ \Psi_n \end{pmatrix}, \]

where \( \Psi_n \) is the wavefunction at the \( n \)th \( z \)-point. By inspection we see that \( m_{n,11} = h^2 f + 2 \), \( m_{n,12} = -1 \), \( m_{n,21} = 1 \) and \( m_{n,22} = 0 \). It is worth noting here that the determinant of each \( M_n \), \( \det(M_n) = 1 \). The product of the \( N \) transfer matrices \( \prod_{n=1}^{N} M_n = M^T \) represents the transformation from \( \Psi_0 \) to \( \Psi_N \). By Bloch’s theorem’s periodic boundary condition, Eq. (3), this transformation can be written

\[ M^T \begin{pmatrix} \Psi_1 \\ \Psi_0 \end{pmatrix} = \begin{pmatrix} m_{11}^T & m_{12}^T \\ m_{21} & m_{22} \end{pmatrix} \begin{pmatrix} \Psi_1 \\ \Psi_0 \end{pmatrix} = \begin{pmatrix} \Psi_{N+1} \\ \Psi_N \end{pmatrix} = e^{ika} \begin{pmatrix} \Psi_1 \\ \Psi_0 \end{pmatrix}. \]

Therefore,

\[ 0 = \det(M^T - e^{ika} \mathbb{I}_2) = (m_{11}^T - e^{ika})(m_{22}^T - e^{ika}) - m_{12}^T m_{21}^T = m_{11}^T m_{22}^T - m_{12}^T m_{21}^T - e^{ika}(m_{11}^T + m_{22}^T) + e^{2ika} = \det(M^T) - e^{ika} \text{Tr}(M^T) + e^{2ika}. \]

Since the determinant of \( M_n \) is 1, then the determinant of any product of \( M_n \), for any \( n \), will also have a determinant of 1, hence \( \det(M^T) = 1 \). Substituting this into Eq. (11) gives

\[ 1 - e^{ika} \text{Tr}(M^T) + e^{2ika} = 0 \]

\[ \text{Tr}(M^T) = e^{ika} + e^{-ika} = 2 \cos(ka) \]

For a given energy, wavevector \( k \) can thus be found by

\[ k = \frac{1}{a} \arccos\left( \frac{1}{2} \text{Tr}(M^T) \right), \]

and the corresponding wavefunction can be found similarly.

III. RESULTS

In the RKKY model of interlayer exchange the oscillations occur as a function of \( k_F d \), where \( k_F \) is the Fermi wave vector and \( d \) is the separation between the two-dimensional magnetic planes. In our model we shall denote \( d_n \) as the width of the non-magnetic layers, corresponding to \( d \) from the RKKY model, and \( d_m \) as the width of the magnetic layers. The length of a GaAs unit cell is labelled \( a_0 \) and has a value of 0.565 nm. We shall also define the average Fermi wave vector \( k_F \) as

\[ k_F = (3\pi^2 \bar{N}_{3D})^{\frac{1}{3}}, \]

corresponding to the Fermi vector \( k_F \) in the ideal RKKY model with a parabolic band. The average 3D carrier concentration \( \bar{N}_{3D} \) is defined as

\[ \bar{N}_{3D} = \frac{1}{d_{n+m}} \int_{\text{unit cell}} N_{3D}(z) \, dz = \frac{N_{2D}}{d_{n+m}}. \]

The superlattice structures being considered in this paper consist of thin (Ga,Mn)As layers interposed with non-magnetic spacer layers. The primary structural parameters that can be changed are the widths of the layers and their composition. Fig. (I(a)) shows the calculated self-consistent charge distribution and potentials for a simple case with a low moment concentration (2%), and the spacer layers are thicker than the magnetic layers. There is a uniform impurity concentration of acceptors throughout the structure, either magnetic, like Mn in the magnetic layers, or from non-magnetic dopants in the spacer layers. The polarisation of carriers is low in this case, and they have an almost totally uniform distribution. These figures show cases where the number of monolayers of non-magnetic layer \( d_m/a_0 = 5 \) and average 3D carrier concentration \( \bar{N}_{3D} = 10^{20} \text{ cm}^{-3} \). The Fermi energy is at \( V_F = 0 \text{ eV} \). The interlayer coupling is in an assumed AFM state, although this may not be the energetically favoured state for such a system.

The effect of increasing the number of moments in the magnetic layers is shown in Fig. (I(b)), where the Mn doping is 8%. As expected, the increased moment concentration increases the size of spin splitting in the magnetic layers, which increases the carrier polarisation. Additionally, the potential due to the magnetic ordering causes a redistribution of carriers to occur, increasing the concentration in the magnetic layer. Another way to cause carrier redistribution is to remove the doping from the non-magnetic layer, which is shown in Fig. (I(c)), where the Mn concentration in the magnetic layers is again 2%. Without a neutralising background charge, Coulomb repulsion opposes carrier redistribution into the non-magnetic layers. The resulting carrier distribution is similar to that of Fig. (I(b)). Although there is a greater concentration of carriers in the magnetic layers, the polarisation is not significantly increased over that seen in Fig. (I(a)) where there is a uniform charge concentration.

To cause stronger confinement of carrier to the magnetic layers, the non-magnetic layers can be made from (Al,Ga)As. The effects of this are shown in Fig. (II(a)), where the carrier concentration in the centre of the magnetic layers is about double that of the centre of the non-magnetic layers. Although there is a slight increase in
FIG. 1: (colour online) The self-consistent charge distribution, $N_{3D}$, and potentials, $V_{\sigma}$, for a double unit cell of three different (Ga,Mn)As/GaAs based superlattice structures in an AFM state. $d_m/\frac{1}{2}a_0=2$, $d_n/\frac{1}{2}a_0=5$ and $N_{3D}=10^{20}$ cm$^{-3}$ in each case. $V_{\sigma}=0$ eV corresponds to the Fermi level. (a) 2% Mn doping and a uniform impurity concentration, (b) 8% Mn doping and a uniform impurity concentration and (c) 2% Mn doping but no impurities in the non-magnetic layer.

the polarisation of carriers over the previous two cases with 2% Mn moment, this is increased significantly when more moments (8%) are put in the magnetic layers, as shown in Fig. 2(b). Because of the strong confinement of carriers to the highly magnetic layers there is a very high polarisation of carriers; this effect is enhanced over that of the 8% case with a doped GaAs spacer which was shown in Fig. 1(b). Finally, Fig. 2(c) shows a case where the non-magnetic layers are thinner than the magnetic layers, and also have a high Mn doping. Following the established trend, carriers are strongly polarised and tightly confined to the magnetic layers. In fact, with an almost total polarisation and a very high depletion of the non-magnetic layers, this structure represents an almost opposite case to that of Fig. 1(a).

Bearing in mind these examples of how changing the structural properties can alter the electronic configuration of the superlattices, the effects of these changes on the IEC will now be explored. This will be done in two parts. Firstly, GaAs based spacers, similar to those shown in Fig. 1 will be considered. The more extreme cases presented in Fig. 2 will be considered in the second half.

A. GaAs spacer

First to be considered is a superlattice structure close to the RKKY limit of infinitely thin magnetic layers surrounded by free unpolarised carriers. For this we shall use thin magnetic layers and a low magnetic moment concentration, as per Fig. 1(a). In Fig. 3(a) the IEC energy, $E_c$, is plotted against the 3D carrier concentration, $\bar{N}_{3D}$, and number of monolayers of GaAs in the non-magnetic spacer, $2d_n/\frac{1}{2}a_0$. The magnetic (Ga,Mn)As layer is 2 monolayers thick and contains 2% Mn local moment doping. There is a uniform acceptor density throughout the structure which gives an average hole concentration of $4.43 \times 10^{20}$ cm$^{-3}$. In this case there are oscillations as a function of both parameters, analogous to the $k_Fd$ oscillations in the ideal quasi one-dimensional RKKY model. For the calculated IEC energy, $E_c$, positive values correspond to FM interlayer coupling being energetically favourable, and negative values correspond to AFM interlayer coupling being the favoured configuration. The RKKY like behaviour observed in Fig. 3(a) is consistent with the results obtained in the tight-binding approach when the exchange coupling energy, $E_c$, is plotted against the two-dimensional carrier concentration, $N_{2D}$, for fixed layer thicknesses. However, it is worth noting that when the exchange coupling is plotted as a function of the non-magnetic spacer thickness,
FIG. 2: (colour online) The self-consistent charge distribution, \(N_{3D}\), and potentials, \(V_\sigma\), for a double unit cell of three different \((\text{Ga,Mn})\text{As}/(\text{Al, Ga})\text{As}\) based superlattice structures in an AFM state. \(d_n/2a_0 = 5\), \(\bar{N}_{3D} = 10^{20}\) cm\(^{-3}\) and the Al concentration is 30% in each case. \(V_\sigma = 0\) eV corresponds to the Fermi level. (a) \(d_m/2a_0 = 2\) and 2% Mn doping (b) \(d_m/2a_0 = 2\) and 8% Mn doping (c) \(d_m/2a_0 = 8\) and 8% Mn doping.

d\(_n\), for a fixed \(N_{2D}\) there are no apparent RKKY oscillations. Because \(N_{3D}\), and therefore \(k_F\), is a function of \(d_n\) these two parameters are not independent when \(N_{2D}\) is fixed. This results in the oscillatory behaviour appearing to be suppressed.

There are, however, real physical reasons for deviation from RKKY behaviour. The data from Fig. 3(a) are replotted in Fig. 5 as a function of \(2\bar{k}_F d_n + 1\). Also plotted is the function

\[
y = \alpha \frac{\sin(x)}{x^2},
\]

where \(\alpha\) is a scaling factor. This function is the asymptotic limit of the pseudo one-dimensional RKKY range function.\(^{17}\) The strength of the interaction is expected to scale with the density of states, and in the 1D case \(\alpha \sim k_F^2\).\(^{18}\) The different series of points on the graph correspond to the series of different non-magnetic spacer thicknesses shown in Fig. 3(a). For a given \(2k_F d_n + 1\), the points with the largest magnitude are those with the greatest \(k_F\); this behaviour is consistent with the expected scaling of \(\alpha\) with \(k_F\). Note the fact that, in order to have improved alignment of the curves, the oscillations were plotted with the effective value of the non-magnetic spacer, \(d_n\), being increased in size by one monolayer, which is denoted \(d_n+1\). This is necessary due to the fact that the magnetic layer in this structure is not infinitely thin, as per the ideal RKKY case, but has a defined width.

Exploring this deviation from RKKY behaviour further, Fig. 3(b) shows the IEC for a superlattice system with a thicker magnetic layer, \(d_m/2a_0 = 8\). All other parameters are as with (a). Examining the AFM peak, the reduction in average carrier concentration of the minimum as the spacer thickness is increased occurs more rapidly, evidenced by the larger derivative of the average 3D carrier density, \(\bar{N}_{3D}\), with respect to the non-magnetic layer thickness, \(d_n\), of the minimum \(E_c\) at low \(d_n\). By way of contrast, at large \(d_n\) this is lower, that is the curve has become much more straight. This is consistent with the effects of large magnetic layers increasing the centre-to-centre distance of the magnetic layers, causing the effect of an apparently larger non-magnetic layer. However, in addition to this, increasing the magnetic layer thickness has introduced additional points of inflection, for reasons that are not immediately obvious.

It is also possible to deviate from RKKY-type behaviour through redistribution of charge. There are two primary methods by which this is achieved, as shown in Figs. 3(b) and (c). The first is that charge is confined to the magnetic layers by the magnetic exchange potential. Fig. 3(a) shows the IEC where the Mn doping has been increased to 8%. However, when the magnetic layer is thin, significant charge redistribution is opposed by the Coulomb potential and the RKKY character is not
FIG. 3: (colour online) The IEC energy, $E_c$, of two (Ga,Mn)As/GaAs based superlattices as a function of the average 3D carrier concentration, $\bar{N}_{3D}$, and the number of monolayers of non-magnetic layer, $2d_n/2a_0$.

Positive (red coloured) values of $E_c$ indicate FM interlayer coupling is energetically favourable and negative (blue coloured) values indicate AFM coupling is favourable. Both superlattices have a 2% Mn doping in the magnetic layer and there is a uniform impurity concentration of acceptors through the structure.

FIG. 4: (colour online) The IEC energy, $E_c$, of two (Ga,Mn)As/GaAs based superlattices as a function of the average 3D carrier concentration, $\bar{N}_{3D}$, and the number of monolayers of non-magnetic layer, $2d_n/2a_0$.

Both superlattices have an 8% Mn doping in the magnetic layer and there is a uniform impurity concentration of acceptors through the structure.

significantly affected. As the figure shows, the main effect is that the size of the IEC is increased. Despite the larger spin splitting and the larger polarisation of carriers caused by the greater moment concentration, the coupling retains an RKKY character.

When the magnetic layer is made wider the increased quantity of magnetic moments now causes additional changes in the oscillatory behaviour, beyond that of simply increasing $d_m$. Fig. 4(b) plots the IEC for a system which now has magnetic layers of 8 monolayers with a Mn doping of 8%. Because of the increased depletion of carriers from the non-magnetic layers, the $N_{3D}$ values at which AFM coupling is expected to occur are now greater for a given non-magnetic layer thickness. Additionally, the damping of the magnitude of the IEC energy with increasing $d_n$ has now significantly changed. While the first FM and AFM maxima are rapidly diminished with increasing non-magnetic spacer, the second FM peak is not greatly affected. The second AFM peak even increases in magnitude with larger $d_n$, and for large spacer it can even be greater than the first.

Note that when the unit cell becomes large and there is a high carrier concentration, the weak coupling and flat minibands make self-consistent convergence difficult; these regions are visible as rough areas on the figures. Such samples would anyway be extremely sensitive to inhomogeneities and fluctuations. No data is shown where...
FIG. 5: (colour online) IEC energy, $E_c$, as a function of $2k_F d_{n+1}$ for a superlattice with magnetic layers with a Mn doping of 2% and 2 monolayer thickness, and a uniform impurity concentration. The (red) curve is an estimate of the ideal RKKY range function.

The calculations have diverged.

The second method of charge redistribution is via a Coulomb potential. Fig. 6(a) shows the IEC profile for a system with a magnetic spacer of two monolayers and a Mn concentration of 2%. However, now there is no neutralising background charge in the non-magnetic layer, so self-consistent redistribution results in the formation of an effective barrier. Fig. 6(c) shows the potentials and charge distribution for a unit cell of this structure in an AFM configuration, again with $d_n/\frac{1}{2}a_0 = 5$ and $N_{3D} = 10^{20}$ cm$^{-3}$. As previously noted, the Coulomb barrier formed is comparable in size to the spin splitting caused by the 2% Mn doping. This results in a similar charge redistribution as in the 8% doped case, although without such strong carrier polarization. As with that case, there is not a significant deviation from RKKY-type behaviour.

Increasing the magnetic spacer thickness now causes more significant changes than seen with the doped spacers. Fig. 6(b) shows the IEC profile for a superlattice with $d_m/\frac{1}{2}a_0 = 8$ with a 2% Mn doping and no impurities in the non-magnetic spacer. In addition to the extra inflection points there is now an additional AFM region. The magnitude of the local minimum in this region does not decrease much with non-magnetic spacer width, and occurs with an almost linear $dN_{3D}/dd_n$. This is now very unlike RKKY behaviour.

To investigate this further we shall now consider superlattice with (Al,Ga)As non-magnetic spacers, so that greater charge redistribution will occur than that caused by the magnetic ordering potential of a high magnetic moment concentration, or the Coulomb potential arising from an undoped spacer.

FIG. 6: (colour online) The IEC energy, $E_c$ of two (Ga,Mn)As/GaAs based superlattices as a function of the average 3D carrier concentration, $N_{3D}$, and the number of monolayers of non-magnetic layer, $2d_n/\frac{1}{2}a_0$.

Both superlattices have a 2% Mn doping in the magnetic layer but there is no charge doping in the non-magnetic spacer layer.

B. (Al,Ga)As spacer

In the previous section it was demonstrated that interlayer coupling in superlattice structures would have an oscillatory behaviour as a function of parameters $N_{3D}$ and $d_n$, analogous to that of RKKY, when the magnetic layers were thin and surrounded by charge. As the structure of the superlattice is changed the IEC would start to deviate from the ideal RKKY behaviour. This is particularly apparent with increased magnetic layer thickness. Changing the 3D charge distribution has a more limited effect; neither large magnetic moment concentration nor a self-consistent Coulomb barrier would cause significant confinement of carriers. In order to investigate these effects further, a band offset will be introduced to fur-
FIG. 7: (colour online) The IEC, $E_c$, of two (Ga,Mn)As/(Al,Ga)As based superlattices as a function of the average 3D carrier concentration, $N_{3D}$, and the number of monolayers of non-magnetic layer, $2d_n/\frac{1}{2}a_0$.

Both superlattices have a 2% Mn doping in the magnetic layer and the non-magnetic layers are (Al$_{0.3}$,Ga$_{0.7}$)As and have no charge doping.

ther confine carriers to the magnetic layers. This will be achieved by using (Al$_{0.3}$,Ga$_{0.7}$)As as the non-magnetic layer material, which has a valence band offset of about 150 meV from GaAs.$^{19,20}$

Fig. 7(a) shows the IEC profile for a structure with a (Ga$_{0.98}$,Mn$_{0.02}$)As magnetic layer of 2 monolayers and an (Al$_{0.3}$,Ga$_{0.7}$)As non-magnetic layer. There is no doping in the non-magnetic layers. The peak FM and AFM coupling strengths are now stronger than in the case with doped GaAs spacers seen in the otherwise identical structure in Fig. 3(a). The charge distribution of this structure is shown in Fig. 2(a), where the barrier confines carriers to the magnetic layers, as expected. However, the $2d_n\frac{k_F}{a_0}$ oscillations are damped more rapidly than with the GaAs spacer, resulting in the second FM and AFM peaks being very weak. This additional damping occurs particularly rapidly with increasing carrier density, $N_{3D}$. As a result, the first anti-ferromagnetic peak barely reduces in magnitude as the non-magnetic layer thickness is increased. Juxtaposing this with the GaAs barrier case, where the largest AFM coupling that can occur when $d_n/\frac{1}{2}a_0 = 10$ is less than a quarter of the size of that when $d_n/\frac{1}{2}a_0 = 2$, we identify this as a significant departure from the previously seen RKKY-like oscillatory behaviour.

Increasing the magnetic moment concentration leads to a more interesting alteration than with the GaAs spacer, where the effect was principally to scale up the magnitude of the IEC energy. Fig. 8(a) shows the IEC for a (Ga,Mn)As/(Al,Ga)As superlattice with an 8% Mn doping in the 2 monolayer magnetic layer, as previously
considered in Fig. 2(b). Now the first AFM peak appears to have two stages. The first is at low spacer thicknesses, where the average hole density at which the maximum occurs decreases with increasing spacer thickness. For large spacer thicknesses the curve has straightened out, and there is almost no dependence on $d_n$ for the sign of the coupling. This characteristic is similar to that exhibited in Figs. 2(b) and 4(b), where the magnetic layer is 8 monolayers thick. This was attributed to loss of independence of the $d_n$ and $N_{3D}$ parameters, as the system became less RKKY-like. Knowing that the band offset and large magnetic ordering cause significant carrier redistribution, particularly, this means that the carrier concentration in the spacer will decrease as a function of spacer thickness. This can account for the weak dependence of $E_c$ on $k_F d_n$. Also, note that the size of the first AFM peak decreases more rapidly at high spacer thicknesses where the average carrier concentration, $N_{3D}$, at which it occurs is not decreasing. This is consistent with the previous observation of enhanced damping with increasing carrier concentration.

With high magnetic layer thicknesses the RKKY-type oscillations have almost completely disappeared. The beating patterns which were emerging in the $d_m / a_0 = 8$ GaAs spacer cases have now come to dominate the IEC. Figs. 7(b) and 8(b) shows this for $d_m / a_0 = 8$, with respectively 2% and 8% Mn doping. In these cases the oscillations occur almost exclusively as a function of hole density, being almost independent of the magnetic layer. Note, however, as was seen in Fig. 2(c), the non-magnetic layer is highly depleted when the magnetic layer is 8 monolayers thick with an 8% Mn doping. This makes computing IEC for larger spacers unfeasible.

IV. DISCUSSION AND RECIPES

Having explored the parameter spaces we will now consider possible structures of a (Ga,Mn)As based superlattice that would exhibit AFM interlayer coupling. Each parameter will be considered for feasibility, and, based on the above calculations, suggestions for values can be made.

The first to be considered is the Mn concentration in the (Ga,Mn)As layers. From the viewpoint of simply creating a viable ferromagnet this is an essential parameter; not only does each substitutional Mn provide a magnetic moment, it also acts as an acceptor and thus this factor controls the hole concentration. Calculations estimate that the minimum hole density for ferromagnetism is $\sim 10^{20}$ cm$^{-3}$. Assuming that each Mn provides one hole, this carrier concentration would correspond to a moment concentration of $\sim 0.5\%$. Experimentally, typical Mn concentrations are in the range of 2-8% ($4.4 \times 10^{20}$ to $1.8 \times 10^{21}$ cm$^{-3}$), which in good quality material could result in higher carrier concentrations than the $10^{19}$ to $10^{21}$ cm$^{-3}$ range considered in these calculations. Whilst the higher magnetic moment concentration can increase the size of the $E_c$ peak, and thus a high moment concentration is favourable, the high carrier concentrations that would be associated with this would cause the strength of the IEC to become extremely weak. This constraint therefore imposes a practical range for Mn concentrations as being between 2 and 4% ($4.4 \times 10^{20}$ to $8.8 \times 10^{20}$ cm$^{-3}$ respectively).

For the non-magnetic spacer thickness the general trend is that the strength of the IEC becomes weaker as the non-magnetic layer becomes thicker. Although this effect is somewhat diminished for the cases where there is strong carrier confinement to the magnetic layers, it is a serious consideration and, ideally, to see strong IEC effects, the non-magnetic layer should be as thin as possible. Furthermore, particularly in cases where the $2k_F d$ behaviour is dominant, as carrier concentration increases the spacer thickness at which the AFM IEC increases inversely. As discussed above, low carrier concentrations are not possible, so therefore it would seem beneficial to make the spacer layers as thin as practicable. Bearing in mind that the average distance between two Mn atoms when the concentration is 3% is of the order of a couple of GaAs unit cells, in order to make the non-magnetic spacer a discernible barrier then 4 monolayers would seem to be a realistic lower bound.

The effect of the magnetic layer thickness on the IEC profile is more subtle, and seems mainly to distort the RKKY behaviour but otherwise, in the limits considered within this study, does not have any negative effects on the interlayer coupling. However, again for interlayer coupling to exist it is necessary that each magnetic layer is itself ferromagnetic. Usually (Ga,Mn)As is grown in bulk layers of many nanometres; the thinnest (Ga,Mn)As epilayers for which published literature exists are 5 nm thick and the (Ga,Mn)As based heterostructures with magnetic layers as thin as 8 monolayers have been reported to be ferromagnetic. It would therefore seem prudent, in order to ensure that the magnetic layers are effective ferromagnets, to prefer to make them thicker. For the 5 mn film some amount of surface depletion should be expected, so a 8 monolayer thick magnetic layer, equivalent to 2.26 nm, is comparable. Of course, if thinner films are shown to be viable then there is no reason not to consider them also.

Based on these constraints, Fig. 9 shows the IEC profile for two candidate superlattices as a function of carrier concentration, $N_{3D}$. Both superlattices are identical in structure except for the composition of the non-magnetic layer. The magnetic layer thickness is 8 monolayers and has a magnetic impurity concentration of $5 \times 10^{20}$ cm$^{-3}$ (2.26%) and the non-magnetic layers are 4 monolayers thick. As expected from the calculations, when the (Al,Ga)As barriers strongly confine carriers to the magnetic layers the IEC energy can have potentially greater magnitudes, although the oscillations have a much higher frequency. In these samples the carrier concentration would be somewhere below the Mn concentration of $5 \times 10^{20}$ cm$^{-3}$ (2.26%), however the exact
multilayers take a typical “worst case” value of the in-plane cubic netocrystalline anisotropic energy of (Ga,Mn)As, we find the energy density of the IEC energy has been observed in EuS/PbS superlattices. Particular interestingly, in a (Ga,Mn)As/(In,Ga)As based superlattice composed of 8 and 4 monolayer width layers respectively two phase transitions were observed. Although there was no evidence that this was due to any AFM effects, this indicates that there is some additional physics at play in these systems, making them of interest for further study. The calculations presented in this paper have shown the importance of the composition of the non-magnetic spacer on the character of the IEC. Using different Al concentrations to tailor the band-offset between layers in a superlattice is a standard tool for designing normal non-magnetic superlattice systems. Utilising this technique in the magnetic superlattices could potentially provide a way to tune to IEC profile to one where AFM coupling is preferential.

Comparing, then, the calculated IEC to the magnetocrystalline anisotropic energy of (Ga,Mn)As, we take a typical “worst case” value of the in-plane cubic anisotropy constant to be of the order of 2000 J m$^{-3}$ at 4.2 K. Using a value of the interlayer coupling energy $E_c = 10 \, \mu J \, m^{-2}$ from Fig. 9 and using the bilayer period of 3.4 nm we find the energy density of the IEC energy is 3000 J m$^{-3}$. Although this is assuming an ideal value of $E_c$, this compares favourably with the anisotropy energy. Furthermore, larger values for the IEC have been found in the tight-binding approach. Therefore, such a superlattice structure might reasonably be expected to be a candidate to exhibit AFM interlayer coupling.

V. CONCLUSION

The composition and structure of (Ga,Mn)As based superlattices can have profound effects on the expected IEC. By examining possible compositions within the broad parameter space that these structures offer it is possible to identify different recipes for devices that might offer the possibility of demonstrating AFM interlayer coupling. Such a study was presented in this paper based on the parabolic band $k \cdot p$ kinetic exchange model. This model ignores spin-orbit and band warping effects, although comparisons to the previously studied microscopic tight-binding model suggest that our results provide a reasonable qualitative or semi-quantitative description of the system. The calculations predict that short period superlattices with magnetic and non-magnetic layers with widths less than 10 monolayers seem be promising candidates. There is existing experimental work in (Ga,Mn)As/GaAs based superlattices with similar dimensions, but this has only exhibited FM IEC. The ideal dimensions suggested by the calculations are therefore feasible and have shown themselves to be viable ferromagnetic DMS material.

Particularly interestingly, in a (Ga,Mn)As/(In,Ga)As based superlattice composed of 8 and 4 monolayer width layers respectively two phase transitions were observed. Although there was no evidence that this was due to any AFM effects, this indicates that there is some additional physics at play in these systems, making them of interest for further study. The calculations presented in this paper have shown the importance of the composition of the non-magnetic spacer on the character of the IEC. Using different Al concentrations to tailor the band-offset between layers in a superlattice is a standard tool for designing normal non-magnetic superlattice systems. Utilising this technique in the magnetic superlattices could potentially provide a way to tune to IEC profile to one where AFM coupling is preferential.

We acknowledge support from EU Grant IST-015728, from UK Grant GR/SB1407/01, from CR Grants KAN400100652, FON/06/E002, AV0Z1010052, and LC510.

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