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Resonant indirect exchange via spatially separated two-dimensional channel

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We apply the resonant indirect exchange interaction theory to explain the ferromagnetic properties of the hybrid heterostructure consisting of a InGaAs-based quantum well (QW) sandwiched between GaAs barriers with spatially separated Mn δ -layer. The experimentally obtained dependence of the Curie temperature on the QW depth exhibits a peak related to the region of resonant indirect exchange. We suggest the theoretical explanation and a fit to this dependence as a result of the two contributions to ferromagnetism—the intralayer contribution and the resonant exchange contribution provided by the QW. © 2015 AIP Publishing LLC.

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Dilute magnetic semiconductors (DMS) have been attracting a lot of attention for quite a while.¹ A lot of efforts have been put forward to combine the numerous advantages of semiconductors with the spin-related phenomena introduced by the magnetic impurities. A special attention nowadays is focused on (Ga, Mn)As DMS which has rather high Curie temperature. However, the details of the mechanism responsible for the ferromagnetism of GaAs doped with a small amount of Mn have not yet been clarified.² It is commonly accepted that it is due to the indirect exchange interaction mediated by the holes. The highest Curie temperature achieved for bulk dilute (Ga,Mn)As samples so far is $T_C \approx 200$ K.³ Mn solubility limit prevents further increase of T_C by increasing the doping level in bulk samples.⁴ In this regard, the GaAs heterostructures with a Mn layer coupled to a spatially separated 2D holes channel have gained a considerable interest.^{5–8} It has been demonstrated that GaAs heterostructure with a Mn δ -layer located in a vicinity of $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum well (QW) shows ferromagnetic behavior similar to that of the bulk (Ga,Mn)As DMS. It was discovered, however, that the ferromagnetic properties depend on the QW depth in a non-monotonic way.⁹ It was suggested that this may be due to falling of the hole bound state at Mn ion into the energy range of occupied 2D heavy holes states in the QW.¹⁰ A theory of the indirect exchange via a spatially separated conducting channel was developed in Refs. 10 and 11. The theory predicted enhancement of the exchange interaction strength due to the resonant tunnel coupling of a bound state at magnetic ion with the continuum of delocalized 2D states in the channel. In this paper, we apply the resonant exchange theory to explain the experimentally observed dependence of the Curie temperature on the QW depth. When discussing 2D structures one should be aware of what is meant by the Curie temperature as there is no spontaneous breaking of a continuous (rotational) symmetry in our

2D Heisenberg ferromagnet. In our theoretical approach, we actually consider the effective exchange constant, i.e., the energy of the indirect exchange interaction between the two neighbouring Mn ions. For 3D case, it is indeed close to the critical Curie temperature for the system to undergo the ferromagnetic phase transition. In 2D case, we define Curie temperature T_C as the one marking the onset of “local ferromagnetic order” when the magnetic correlation length well exceeds the distance between the Mn ions. In the experiment so defined T_C is obtained from a hump on the in-plane electrical resistance temperature dependence,^{12,13} a more detailed discussion on the critical temperature in this system is given in Refs. 14 and 15.

We consider the problem of indirect exchange between two magnetic ions i and j via 2D channel as shown schematically in Fig. 1(a). Here, I_{ij} is the spin projection of the i th(j th) magnetic ion, $\varepsilon_{i,j}$ is the ion’s bound state energy, and $d_{i,j}$ is the distance between the ion and the channel. Indirect exchange mediated by free carriers is usually described on the basis of Ruderman-Kittel-Kasuya-Yosida (RKKY) theory.¹⁶ Separation of the magnetic ions from the free carriers gas by a potential barrier leads to the suppression of the effect due to decay of the 2D carriers wavefunction into the region containing magnetic centers. However, if a magnetic

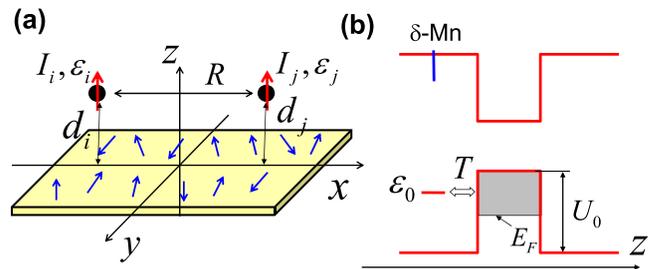


FIG. 1. Illustration of the indirect exchange interaction via 2D channel (a) and band diagram of a InGaAs-based heterostructure with a QW and Mn δ -layer (b).

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ion possess a bound state with an energy within the 2D gas energy spectra, a resonant tunneling may occur. Such a resonant coupling cannot be straightforwardly accounted for within the RKKY approach. We consider two magnetic ions i and j coupled to the free electron gas in the tunneling Hamiltonian formalism

$$\begin{aligned} H &= H_0 + H_T + H_J, \\ H_0 &= \varepsilon_i a_i^\dagger a_i + \varepsilon_j a_j^\dagger a_j + \int \varepsilon_\lambda c_\lambda^\dagger c_\lambda d\lambda, \\ H_T &= \int (t_{i\lambda} a_i^\dagger c_\lambda + t_{j\lambda} a_j^\dagger c_\lambda + h.c.) d\lambda, \\ H_J &= JA(I_i s a_i^\dagger a_i + I_j s a_j^\dagger a_j), \end{aligned} \quad (1)$$

where H_0 is the Hamiltonian of the system without tunnel coupling and spin-spin interaction, H_T is the tunnelling term, H_J is the exchange term, a_i^\dagger and a_i are the creation and annihilation operators for the bound states at the impurity ion i , characterized by the energy ε_i and wavefunction ψ_i , c_λ^\dagger and c_λ are the creation and annihilation operators for a continuum state characterized by the quantum number(s) λ , having the energy ε_λ and the wavefunction φ_λ , J is the exchange constant, A is the wavefunction amplitude at the ion's site squared, s is the electron spin projection, and $t_{i,\lambda}$ is the tunneling matrix element¹⁰

$$t_i(k) = \sqrt{\frac{\hbar^2 T_i}{2\pi m}} e^{i\mathbf{k}\mathbf{R}_i}, \quad (2)$$

where m is the in-plane effective mass and T_i is the energy parameter for the tunneling

$$T_i = \alpha U_0 e^{-2qd_i}, \quad (3)$$

where U_0 is the height of the potential barrier separating the magnetic centers from the channel, $q = \sqrt{2m_\perp U_0}/\hbar$, m_\perp is an effective mass along the tunneling direction, the dimensionless parameter α depends on the channel and magnetic centers details.¹¹ We obtained the exchange energy between the two ions in the form^{10,17}

$$E_{ij} = \frac{1}{\pi} \int_0^{E_F} d\varepsilon \arctan \left[\frac{8\pi^2 j^2 T_i T_j J_0(kR) Y_0(kR)}{((\varepsilon_i - \varepsilon)^2 - j^2)((\varepsilon_j - \varepsilon)^2 - j^2)} \right], \quad (4)$$

where $j = JA|I||s|$, E_F is the Fermi level of the carriers in the channel (zero temperature is assumed), $k = \sqrt{2m\varepsilon}/\hbar$, and J_0 and Y_0 are Bessel and Neumann functions of zeroth order.

Formula (4) is valid for both resonant and non-resonant tunnel coupling. The resonant case corresponds to the bound states energy levels lying within the energy range of the occupied states in the 2D channel

$$\varepsilon_i, \varepsilon_j \in [0, E_F]. \quad (5)$$

In this regime, the main contribution to the exchange energy (4) comes from the poles of the arctan argument and can be estimated as

$$\begin{aligned} E_{ij} &\approx \gamma \sqrt{jT}, & \text{if } \beta > 1 \\ E_{ij} &\approx \gamma \beta \sqrt{jT}, & \text{if } \beta < 1 \\ \beta &= \frac{\sqrt{jT}}{|\varepsilon_i - \varepsilon_j|}, & T = \sqrt{T_i T_j}. \end{aligned} \quad (6)$$

Here, γ is given by

$$\gamma = \sqrt{2\pi} [J_0(k_i R_{ij}) Y_0(k_i R_{ij}) + J_0(k_j R_{ij}) Y_0(k_j R_{ij})]^{1/4}, \quad (7)$$

where R_{ij} is the distance between the ions. Unlike RKKY theory, here the Fermi wavenumber k_F is replaced by the "resonant" wavenumbers corresponding to the bound levels: $k_i = \sqrt{2m\varepsilon_i}/\hbar$. For the experimental situation considered below the parameter γ appears to be $\gamma \approx 1$ being still far from the first maximum of the oscillations. Approximation (6) is quite good as illustrated in Fig. 2. The dotted curve shows the exchange energy calculated according to (4) for $\varepsilon_i = \varepsilon_j = \varepsilon_0$, while the solid curve shows the approximation (6) assuming $\gamma = 1$. The exchange energy in the resonant case is much larger than in the non-resonant one. The latter case agrees with the RKKY approach. Indeed, the integration (4) for the case when the arctan argument has no poles and therefore is small while the tunneling is weak yields¹⁰

$$\begin{aligned} E_{nr} &= \frac{8\pi T^2 j^2 E_F}{\varepsilon_0^4} \chi(R), \\ \chi(R) &= J_0(k_F R) Y_0(k_F R) + J_1(k_F R) Y_1(k_F R), \end{aligned} \quad (8)$$

where R is the mean distance between the ions. Note that the resonant and non-resonant cases have different parametric dependence on the tunneling parameter T and the exchange parameter j , this leads to substantial amplification of the indirect exchange in the resonant case.

We applied the theory of the resonant exchange to the hybrid GaAs/Mn δ -layer/GaAs/In_xGa_{1-x}As/GaAs heterostructure, which was also studied experimentally. The energy diagram for this system is shown schematically in Fig. 1(b). The Mn content is 0.25–0.3 monolayers (ML), the distance between the Mn layer and the In_xGa_{1-x}As QW is $d = 3$ nm

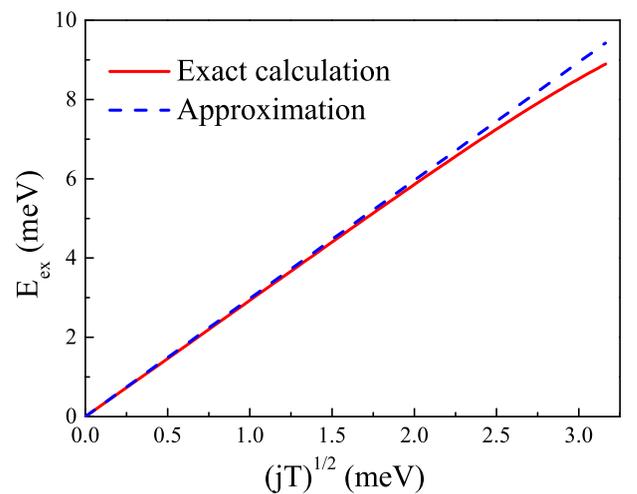


FIG. 2. Approximation for indirect resonant exchange energy (dashed line) compared to exact result (solid line).

(at that the mobility of the 2D holes in the QW remains almost unaffected by the Mn layer¹⁸), the thickness of the QW is 10 nm and its depth is controlled by In concentration x . For the detailed description of the structure see Ref. 8. Due to the Mn diffusion the Mn layer thickness appears to be as large as ≈ 3 nm,¹⁹ it is thus natural to expect that the ferromagnetic properties of such a layer would resemble the bulk ferromagnetism of (Ga,Mn)As. Indeed, the ferromagnetism in such a layer alone has been confirmed experimentally,²⁰ its ferromagnetic properties have been also studied theoretically.²¹ However, the samples containing a QW in addition to the Mn layer were shown to exhibit ferromagnetic properties, which were non-monotonously dependent on the QW depth.⁹ The dependence of Curie temperature on the parameters of the QW suggests that the indirect exchange interaction is partly due to 2D holes sitting in the QW. Another explanation of the observed phenomena could be the redistribution of the holes between Mn layer and the QW. At that the non-monotonous dependence of Curie temperature on holes concentration might be attributed to the impurity band formed in (Ga, Mn)As. The optimal filling factor for the impurity band holes hopping is 0.5, so by sweeping the filling factor from 1 down to 0 one would expect the ferromagnetism to exhibit non-monotonous behavior with maximum at the half-filled impurity band.²² This explanation was, however, criticized²³ pointing out that no non-monotonous behavior is observed if As-antisite defects are properly taken into account.²⁴ Moreover, such scenarios imply radical change of the filling factor, i.e., the concentration of holes going to the QW should exceed that of holes residing in the Mn layer. The holes concentration in the QW in our samples never exceeds $2.3 \times 10^{12} \text{ cm}^{-2}$ while in the Mn layer (with Mn content 0.25–0.3 ML) it is not less than 10^{13} cm^{-2} according to our measurements (also confirmed in Ref. 20). Thus, the hole concentration in the QW is too small to explain the change of the Curie temperature and its non-monotonous behavior by this mechanism. At the same time, it was proved experimentally that the holes in the QW are spin polarized due to strong tunnel coupling with Mn,^{15,18,25} thus one would naturally expect their participation in the indirect exchange between Mn ions. This interaction must be considered with account for the resonant coupling as described above. Indeed, the acceptor binding energy of Mn in GaAs is comparable to the QW depth for the holes. This makes it possible to meet the condition (5) as shown in Fig. 1(b), where ε_0 denotes the average energy of the bound state of a hole at Mn (zero energy corresponds to the first heavy holes quantization level in the QW). We made a fit to the experimental data⁹ (circles in Fig. 3) assuming that there are two contributions to the indirect exchange interaction between the Mn ions. The first one is assumed to be itinerant ferromagnetism of the Mn δ -layer itself mediated by weakly localized holes located in the layer in the same manner as in the bulk (Ga, Mn)As. This contribution does not depend on the QW properties. The second contribution is the resonant indirect exchange via the 2D holes of the QW. This one is treated on the basis of our theoretical result (4). We demonstrate that the maximum of the Curie temperature corresponds to the resonant indirect exchange via the 2D holes of the QW, while its decrease for both too shallow and too deep

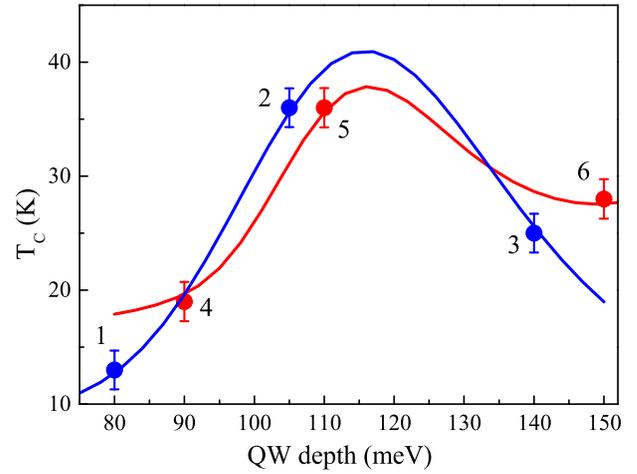


FIG. 3. Dependence of the Curie temperature on the QW depth. Experiment (circles) and theoretical fit (solid curves).

QW is explained by driving the system out of the resonance (5). In our calculation, we assumed the Curie temperature being the sum of the two terms

$$T_C = T_{C1} + T_{C2}, \quad (9)$$

where T_{C1} does not depend on the QW properties. In calculation of the second term T_{C2} , we assumed that the energy levels at Mn ions are normally distributed having an average value ε_0 and dispersion σ_ε . The distance between the neighbouring ions was assumed to be constant, equal to the mean one R . For the Mn δ -doping of 0.3 ML, one can take $R = 1.5$ nm. We checked that taking into account some distribution of the distances as well as varying the mean value has little effect on the resonant exchange term. This is because γ defined by (7) is a very weak function of R in the vicinity of $\gamma = 1$. On the contrary, the Mn bound levels energy distribution does play an important role and must be accounted for. In order to introduce the energy levels distribution into the fitting procedure, it is convenient to replace the approximation (6) by a similar function

$$\tilde{E}(\varepsilon_i, \varepsilon_j) = \frac{\gamma^2 jT}{|\varepsilon_i - \varepsilon_j| + \gamma\sqrt{jT}}. \quad (10)$$

The resonant contribution to the Curie temperature T_{C2} (9) is calculated using the following expression:

$$T_{C2} = \frac{2}{k_B} \int_0^{E_F} d\varepsilon \int_{-\infty}^{+\infty} d\varepsilon' P(\varepsilon)P(\varepsilon')\tilde{E}(\varepsilon, \varepsilon'), \quad (11)$$

where

$$P(\varepsilon) = \frac{1}{\sqrt{2\pi}\sigma_\varepsilon} e^{-\frac{(\varepsilon-\varepsilon_0)^2}{2\sigma_\varepsilon^2}}, \quad (12)$$

k_B is the Boltzmann constant. In the limiting case of a δ -like distribution $\sigma_\varepsilon \rightarrow 0$ expression (11) yields

$$T_{C2} = \begin{cases} \frac{2}{k_B} \gamma\sqrt{jT}, & \varepsilon_0 \in [0, E_F] \\ 0, & \text{otherwise,} \end{cases} \quad (13)$$

TABLE I. The parameters of the fit.

No.	T_C , K	U_0 , meV	p , cm^{-2}	E_F , meV	T_{C1} , K	\sqrt{jT} , meV	σ_e , meV
1	13	80	5.6×10^{11}	7.8	9	3.3	18
2	36	105	8.9×10^{11}	12.5	9	3.3	18
3	25	140	1.8×10^{12}	25.2	9	3.3	18
4	19	90	0.7×10^{11}	1.0	17	4.1	13
5	36	110	3.0×10^{11}	4.2	17	4.1	13
6	28	150	2.3×10^{12}	32.2	17	4.1	13

i.e., the resonant contribution vanishes whenever ε_0 goes below or above the energy range occupied by the carriers in the QW. The approach was used to fit the experimental values of the Curie temperature measured for two series of samples. The samples 1–3 had 0.25 ML of Mn and the QW depth for the holes U_0 varied from 80 to 140 meV, the samples 4–6 had 0.3 ML of Mn with U_0 varied from 90 to 150 meV. The fits for the two series of experimental points are presented in Fig. 3. The average value of the Mn bound state for the best fit was $\varepsilon_0 = U_0 - 103$ meV, i.e., 103 meV above the top of the valence band for GaAs, which roughly matches the Mn acceptor binding energy (≈ 110 meV). This value was the same for the two fits. The hole concentration (and their Fermi level) was derived from the transport experiments^{8,18} and the QW depth was obtained from the optical experiments. These values are given in Table I along with the other parameters of the fit. Other fitting parameters were: non-resonant component of the Curie temperature T_{C1} , bound state energy dispersion σ_e , and the product jT . As it is seen from Table I, for samples 4–6 T_{C1} is higher than for the samples 1–3, this is consistent with the latter having weaker Mn doping. The value of T_{C1} appears to be somewhat smaller than that reported for similar samples having no QW.²⁰ However, we extracted the Curie temperature from the derivative of sheet resistance temperature dependence dR/dT as suggested in Ref. 13 rather than from $R(T)$ as done in Ref. 20. Taking this into account T_{C1} appears to be close to that measured for single Mn layer. The larger jT product for the more heavily doped samples can be understood if we recall that Mn layer has a finite thickness, expected to be larger for larger Mn concentration due to Mn diffusion.⁸ Thus, the minimal distance between the Mn layer and the QW is decreased with increase of Mn doping level resulting in increase of the tunneling parameter. The difference in the energy levels dispersion σ_e perhaps cannot be unambiguously explained with the similar plain arguments. What is read from the fit (Table I) is that the diagonal disorder is somewhat smaller in the Mn layer with higher Mn concentration. Overall, the fit shows good agreement with the experimental data and the values of the fitting parameters seem quite reasonable. It might be also useful to illustrate the interplay between the two contributions to the ferromagnetism. Shown in Fig. 4 are the two contributions as a function of the spacer thickness d . For the calculation presented in Fig. 4, we took the parameters of sample 5 with extrapolation to different spacer thickness. The intralayer contribution (T_{C1} term) does not depend on d , while the QW contribution depends on d via the tunneling parameter. For the resonant case, this dependence is weaker than for the non-resonant one as T_C roughly follows the \sqrt{T} dependence (6) rather than T^2 (8). Despite that

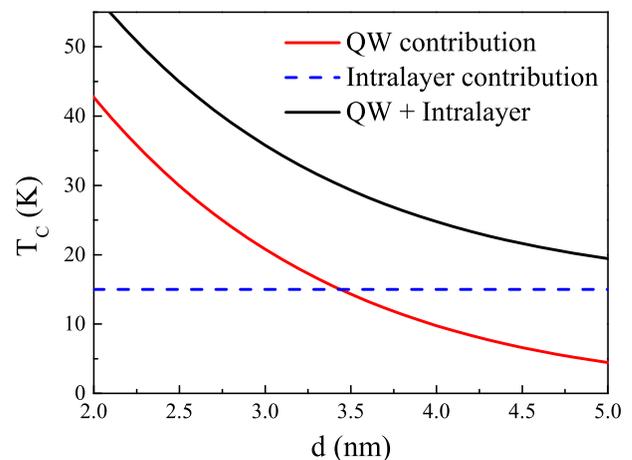


FIG. 4. The calculated dependence of the Curie temperature on the distance between QW and Mn δ -layer.

the dependence of $T_C(d)$ appears to be still stronger than observed in an experiment.⁹ We attribute this disagreement to the uncertainty in determination of d as it is comparable to the thickness of the Mn layer itself (≈ 3 nm). The detailed analysis here requires more experimental data.

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