Spin-orbit mediated anisotropic spin interaction in interacting electron systems

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We investigate interactions between spins of strongly correlated electrons subject to the spin-orbit interaction. Our main finding is that of a novel, spin-orbit mediated anisotropic spin-spin coupling of the van der Waals type. Unlike the standard exchange, this interaction does not require the wave functions to overlap. We argue that this ferromagnetic interaction is important in the Wigner crystal state where the exchange processes are severely suppressed. We also comment on the anisotropy of the exchange between spins mediated by the spin-orbital coupling.

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Introduction. Studies of exchange interaction between localized electrons constitutes one of the oldest topics in quantum mechanics. Strong current interest in the possibility to control and manipulate spin states of quantum dots has placed this topic in the center of spintronics and quantum computation research. As is known from the papers of Dzyaloshinskii [1] and Moriya [2], in the presence of the spin-orbital interaction (SOI) the exchange is anisotropic in spin space.

Being a manifestation of quantum tunneling, the exchange is exponentially sensitive to the distance between electrons [3]. This smallness of the exchange leads to a large spin entropy of the Wigner crystal state, as compared to the Fermi liquid state, of diluted two-dimensional electron gas in semiconductor field-effect transistors [4]. The consequence of this, known as the Pomeranchuk effect, is spectacular: Wigner crystal phase is stabilized by a finite temperature.

In this work we show that when subjected to the spin-orbit interaction, as appropriate for the structure-asymmetric heterostructures and surfaces [5], interacting electrons acquire a novel non-exchange coupling between the spins. The mechanism of this coupling is very similar to that of the well-known van der Waals (vdW) interaction between neutral atoms. This anisotropic interaction is of the ferromagnetic Ising type. It lifts extensive spin degeneracy of the Wigner crystal and leads to the long-range ferromagnetic order. We also re-visit and clarify the role of spin-orbit interaction in lowering the symmetry of the exchange coupling between spins. Particularly, we point out that the exchange Hamiltonian, despite its anisotropic appearance, retains spin-rotational invariance to the second order in the spin-orbital coupling. We argue that spin-rotational symmetry is broken only in the forth order in SOI coupling.

Calculation of the vdW coupling. To illuminate the origin of the vdW coupling, we consider the toy problem of two single-electron quantum dots described by the double well potential [6, 7], see Figure 1.

\[ V(x_j, y_j) = \frac{m \omega_x^2}{2a^2} (x_j^2 - a^2/4)^2 + \frac{m \omega_y^2}{2} y_j^2, \]  

(1)

where the \( \omega_{x/y} \) are confinement frequencies along \( x/y \) directions. The electrons, indexed by \( j = 1, 2 \), are subject to SOI of the Rashba type [6] with coupling \( \alpha_R \)

\[ H_{SO} = \sum_{j=1,2} \alpha_R \vec{p}_j \times \vec{\sigma}_j \cdot \hat{z}, \]  

(2)

where \( \vec{\sigma}_j \) are the Pauli matrices and \( \hat{z} \) is normal to the plane of motion. Finally, electrons experience mutual Coulomb repulsion so that the total Hamiltonian reads

\[ H = \sum_{j=1,2} \left[ \frac{p_j^2}{2m} + V(x_j, y_j) \right] + \frac{e^2}{|\vec{r}_1 - \vec{r}_2|} + H_{SO}. \]  

(3)

At large separation between the two dots the exchange is exponentially suppressed and the electrons can be treated as distinguishable particles. One then expects that Coulomb-induced correlations in the orbital motion of the electrons in two dots translate, via the spin-orbit interaction, into correlation between their spins. Consider the distance between the dots, \( a \), much greater than the typical spread of the electron wave functions, \( 1/\sqrt{m \omega_x} \). In this limit the electrons are centered about different wells, and the potential can be approximated as

\[ V(\vec{r}_1, \vec{r}_2) \approx \frac{1}{2} m \omega_x^2 ((x_1 - a/2)^2 + (x_2 + a/2)^2) + \frac{1}{2} m \omega_y^2 (y_1^2 + y_2^2). \]  

(4)
At this stage it is crucial to perform a unitary transformation which removes the linear spin-orbit term from \( \mathcal{H}_{\text{SO}} \) which results in higher order in the Rashba coupling \( \alpha_R \) contributions as given by \( \mathcal{H} = U \mathcal{H} U^\dagger \) below:

\[
\tilde{\mathcal{H}}_{\text{SO}} = \sum_{j=1,2} \left[ -\alpha_R^2 \tilde{L}_j^z \tilde{\sigma}_j^x + \frac{4}{3} \alpha_R^2 (y_j \tilde{\sigma}_j^y + x_j \tilde{\sigma}_j^x) \tilde{L}_j^z \right] + \frac{2}{3} \alpha_R^2 \left( y_j \tilde{\sigma}_j^x - x_j \tilde{\sigma}_j^y \right) + O(\alpha_R^4) \tag{6}
\]

Here \( \tilde{L}_j^z \) is the angular momentum of the \( j^{th} \) electron, \( \tilde{L}^z = \hat{p}_y - y \hat{p}_z \), and tilde denotes unitarily rotated operators. The calculation is easiest when the confining energy is much greater than both the Coulomb energy \( e^2/a \) and the spin-orbit energy scale \( \sqrt{m\omega_R} \). In terms of the new (primed) coordinates \( r'_1 = r_1 - \tilde{a}/2 \) and \( r'_2 = r_2 + \tilde{a}/2 \) centered about \((a/2,0)\) and \((-a/2,0)\), respectively, the interaction potential \( e^2/|r_1^2 - r_2^2 + \tilde{a}| \) is expanded in powers of \( 1/\tilde{a} \) keeping terms up to second order in the dimensionless relative distance \( (r_1^2 - r_2^2 + \tilde{a})/\tilde{a} \). The linear term, \( e^2 (x'_1 - x'_2)/a^2 \), slightly renormalizes the equilibrium distance between the electrons and can be dropped from further considerations. In terms of symmetric (S) and anti-symmetric (A) coordinates:

\[
x_{S/A} = \frac{x'_1 \pm x'_2}{\sqrt{2}}; \quad y_{S/A} = \frac{y'_1 \pm y'_2}{\sqrt{2}} \tag{7}
\]

the quadratic term \( e^2 (2(x'_1 - x'_2)^2 - (y'_1 - y'_2)^2)/2a^4 \) renormalizes the anti-symmetric frequency \( \omega_{SA}^2 = \omega^2 + 4e^2/(ma^2) \) and \( \omega_{AY}^2 = \omega^2 + 2e^2/(ma^2) \) while leaving the symmetric ones unmodified, \( \omega_{Sx}^2 = \omega_x^2 \) and \( \omega_{Sy}^2 = \omega_y^2 \). Quite similarly to the textbook calculation of the vdW force, the resulting Hamiltonian \( \tilde{\mathcal{H}} = \tilde{\mathcal{H}}_S + \tilde{\mathcal{H}}_A + \tilde{\mathcal{H}}_{SO} \) becomes that of harmonic oscillators

\[
\tilde{\mathcal{H}}_{S/A} = \frac{\tilde{p}_{S/A}^2}{2m} + \frac{m}{2} (\omega_{S/A}^2 \tilde{x}_{S/A}^2 + \omega_{S/A}^2 \tilde{y}_{S/A}^2) \tag{8}
\]

perturbed by \( \tilde{\mathcal{H}}_{SO} = \tilde{\mathcal{H}}_{SO}^{(2)} + \delta \tilde{\mathcal{H}}_{SO}^{(2)} + O(\alpha_R^3) \), where

\[
\tilde{\mathcal{H}}_{SO}^{(2)} = \frac{-m \alpha_R^2}{2} \left[ (x_S \tilde{p}_y S - y_S \tilde{p}_x S) + S \leftrightarrow A \right] (\tilde{\sigma}_1^x + \tilde{\sigma}_2^x) - \frac{m \alpha_R^2}{2} \left[ y_S \tilde{p}_y A - y_A \tilde{p}_x S \right] \leftrightarrow A (\tilde{\sigma}_1^x - \tilde{\sigma}_2^x) \tag{9}
\]

\[
\delta \tilde{\mathcal{H}}_{SO}^{(2)} = \frac{n \alpha_R}{2} [y_S \tilde{p}_y (\tilde{\sigma}_1^x - \tilde{\sigma}_2^x) + \tilde{p}_y A (\tilde{\sigma}_1^x + \tilde{\sigma}_2^x)]. \tag{10}
\]

It is evident from Eqn. \( \text{(9,10)} \) that the leading corrections to the ground state energy is obtained either by the excitation of a single \( y \)-oscillator (through \( \text{Eqn. (10)} \)) and by the simultaneous excitation of oscillators in both the \( x \) and \( y \) directions (through \( \text{Eqn. (11)} \))

\[
\Delta E = \sum_{i,j=S,A} \left| \frac{|0 \delta \tilde{H}_{SO}^{(2)} |y_i|}{\omega_{iy}} \right|^2 + \left| \frac{|0 \delta \tilde{H}_{SO}^{(2)} |x_i, y_j|}{\omega_{ix} + \omega_{iy}} \right|^2 \tag{11}
\]

It is easy to see that the spin-dependent contributions from \( \delta \tilde{H}_{SO}^{(2)} \) cancel exactly while those originating from \( \tilde{H}_{SO}^{(2)} \) do not, resulting in the novel spin interaction

\[
H_{vdW} = \frac{1}{8} \frac{m \alpha_R^4}{4 \alpha_R^2} (\tilde{\sigma}_1^x \tilde{\sigma}_2^x \left( \phi (\omega_{Sy}, \omega_{Sx}) + \phi (\omega_{Ay}, \omega_{Ax}) \right) - \phi (\omega_{Ay}, \omega_{Sx}) - \phi (\omega_{Sy}, \omega_{Ax})), \tag{12}
\]

where the function \( \phi \) is given by a simple expression

\[
\phi (x, y) = \frac{(x - y)^2}{xy (x + y)}. \tag{12}
\]

In case of cylindrically symmetric dots, \( \omega_x = \omega_y \),

\[
H_{vdW} = - \frac{\alpha_R^2}{2} \frac{m \alpha_R^4}{4 \alpha_R^2} (\tilde{\sigma}_1^x \tilde{\sigma}_2^x). \tag{13}
\]

The physics of this novel interaction is straightforward: it comes from the interaction-induced correlation of the orbital motion of the two particles, which, in turn, induces correlations between their spins via the spin-orbit coupling. The net Ising interaction would have been zero if not for the shift in frequency of the anti-symmetric mode due to the Coulomb interaction. Note that the coupling strength exhibits the same power-law decay with distance as the standard van der Waals interaction.

From Eqn. \( \text{(11)} \), it follows that in the extreme anisotropic limit of \( \omega_y \rightarrow \infty \), or equivalently, the one-dimensional (1D) limit, there is no coupling between spins. This result is understood by noting that 1D version of SOI, given by \( \alpha_R \sum_j \sigma_j^y \sigma_{j+1}^y \), can be gauged away to all orders in \( \alpha_R \) by a unitary transformation \( U_{1D} = \exp[im \alpha_R (x_1 \sigma_1^y + x_2 \sigma_2^y)] \). Hence the absence of the spin-spin coupling in this limit. However, either by including magnetic field (Zeeman interaction, see below) in a direction different from \( \sigma^y \), or by increasing the dimensionality of the dots by reducing the anisotropy of the confining potential, the spin-orbital Hamiltonian acquires additional non-commuting spin operators. The presence of the mutually non-commuting spin operators (for example, \( \sigma^x \) and \( \sigma^y \) in \( \text{(2)} \)) makes it impossible to gauge the SOI completely, opening the possibility of fluctuation-generated coupling between distant spins, as in equation \( \text{(13)} \).

Effect of the magnetic field. For simplicity, we neglect orbital effects and concentrate on the Zeeman coupling, \( \mathcal{H}_z = - \Delta \sum_j \sigma_j^z/2 \), where \( \Delta = g \mu_B \). Unitary transformation \( \text{(5)} \) changes it to \( H_z = - \Delta \sum_j \alpha_R \sigma_j^z/2 + \delta \tilde{\mathcal{H}}_z \).

\[
\delta \tilde{\mathcal{H}}_z = - \sum_{j=1,2} \alpha_R \Delta \left( x_j \tilde{\sigma}_j^x + y_j \tilde{\sigma}_j^y \right) \tag{14}
\]
describes the coupling between the Zeeman and Rashba terms. In the basis (7) it reduces to
\[
\delta \hat{H}_Z = -m \Delta_2 \alpha_R \frac{ys(\sigma^y_1 + \sigma^y_2)}{\sqrt{2}} + x_s(\sigma^x_1 + \sigma^x_2) + \frac{m \omega_{0R}}{\sqrt{2}},
\]
(15)
For sufficiently strong magnetic field, \(\Delta_2 \gg \sqrt{m \omega_{0R}}\), \(\hat{H}_{SO}\) can be neglected in comparison with \(\delta \hat{H}_Z\). Calculating second order correction to the ground state energy of the two dots, represented as before by \(\hat{H}_S + \hat{H}_A\), and extracting the spin-dependent contribution, we obtain
\[
\Delta E_Z = -\Delta_2 \alpha_R \frac{y_s}{a} (2 \frac{\sigma^x_1 \sigma^x_2}{\omega^2} - \frac{\sigma^y_1 \sigma^y_2}{\omega^2}).
\]
(16)
In the extreme anisotropic limit \(\omega_y \to \infty\) the dots become 1D and we recover the result of Ref. [11]. For the isotropic limit \(\omega_x = \omega_y\), the coupling of spins acquires a magnetic dipolar structure identical to that found in Ref. [12].

Anisotropy of the exchange. Next, we allow for the electron tunneling between the dots. The spin dynamics of the electrons is now described by the sum of exchange and the van der Waals interactions, \(H = H_{Ex} + H_{vdW}\). Here the exchange coupling, \(H_{Ex}\), contains both isotropic and possible anisotropic interactions, while \(H_{vdW}\) is given by (14) and (15). In the absence of spin-orbit interaction, the total spin is conserved and the Hamiltonian is SU(2) invariant. As such, the only spin interaction allowed has the well known isotropic form \(H_{Ex} \sim \sigma_1 \cdot \sigma_2\). The anisotropy of the exchange is mediated by the spin-rotational symmetry breaking SOI (2). When the tunneling is no longer spin-conserving, electron spins precess while exchanging their respective positions, giving rise to the anisotropic terms. As a result [13, 14, 15]
\[
H_{Ex} = \frac{J}{4} \left( b \sigma_1 \cdot \sigma_2 + D d \cdot \sigma_1 \times \sigma_2 + \Gamma (\hat{d} \cdot \sigma_1)(\hat{d} \cdot \sigma_2) \right),
\]
(17)
where \(\hat{d}\) is the unit Dzyaloshinskii-Moriya vector, of amplitude \(D\), with odd dependence on the spin-orbit coupling \(\alpha_R\). Coefficients \(b\) and \(\Gamma\) have even dependence on the spin-orbit coupling \(\alpha_R\) and \(\Gamma\), while the exchange integral \(J\), independent of \(\alpha_R\) in this representation, sets the overall energy scale. The direction of the DM vector can be understood as follows. As the \(D\)-term must be even under exchange operation \(P: 1 \leftrightarrow 2\), its amplitude must be odd with respect to inter-spin distance \(\hat{d} = \vec{r}_1 - \vec{r}_2 = a \vec{x}\), hence \(\hat{d} \sim \hat{a} = \hat{y}\). In addition, as \(\hat{z} \rightarrow -\hat{z}\) transformation in (2) changes sign of \(\alpha_R\), it must be that \(\hat{d} \sim \vec{z}\) as well. Thus, it must be that \(\hat{d} = \vec{y} \times \vec{a} = \hat{y}\).

In the simplest approximation one neglects the “remnants” of SOI [6] altogether and writes the only possible exchange coupling \(H_{Ex}^{(0)} = \frac{J}{2} \sigma_1 \cdot \sigma_2\) in terms of unitarily transformed spin operators \(\tilde{\sigma}_j\). The meaning of this interaction is understood in the original basis by undoing the unitary transformation, \(H_{Ex}^{(0)} = U \tilde{H}_{Ex}^{(0)} U\). Using (3) and replacing \(\vec{r}_1, \vec{r}_2\) by their respective average values, \(a/2 \hat{x}\) and \(-a/2 \hat{x}\), one observes that spin 1 (2) is rotated about \(\hat{y}\) axis by the angle \(\theta = m \omega_{0a}\) in clockwise (counterclockwise) direction. As a result, one immediately obtains Eq. (17) with parameters
\[
b_0 = \cos 2\theta, D_0 = \sin 2\theta, \Gamma_0 = 1 - \cos 2\theta, \hat{d} = \hat{y}.
\]
(18)
As it originated from the SU(2)-invariant scalar product \(\vec{S}_1 \cdot \vec{S}_2\), the Hamiltonian (17) with parameters (18) does not break spin-rotational SU(2) symmetry, despite its asymmetric appearance. Because of its “non-diagonal” nature, the \(D\)-term affects the eigenvalues only in \(D^2 \sim \theta^2\) order, and must always be considered together with the \(\Gamma\)-term. In the current situation (18), the two contributions compensate each other exactly. This important observation, made in Ref. [14] (see also [17]), was overlooked in several recent calculations of the DM term [15, 18, 19].

It is thus clear that the symmetry-breaking DM term must originate from so far omitted \(\hat{H}_{vdW}\). To capture it, we set up the exchange problem calculation along the lines of the standard Heitler-London (HL) approach. Despite its well-known shortcomings [20, 21, 22], this approach offers conceptually simple way to estimate exchange splitting [2] and the structure of anisotropic spin coupling. Our basis set is formed by the antisymmetrized two-particle wave function \(|\psi\rangle = |\psi\rangle - |\overline{\psi}\rangle\),
\[
|\psi\rangle = \varphi(1, 2)(c_1 \uparrow 1 \downarrow) + c_2 \uparrow \downarrow) + c_3 \downarrow \uparrow) + c_4 \downarrow \downarrow)
\]
(19)
is written in terms of unknown coefficients \(c_{1-4}\). Here \(\varphi(1, 2) = f(x_1 - a/2)f(y_1)f(x_2 + a/2)f(y_2)\) describes spatial wave function of distinguishable particles localized near \((a/2, 0)\) and \((-a/2, 0)\), respectively, and \(f(x-x_0)\) denotes the ground state wave function of one-dimensional harmonic oscillator centered around \(x = x_0\). As constructed, \(\varphi(1, 2)\) is the lowest energy eigenstate of two particles moving in the potential profile [4].

The rest of the confining potential, Eq. (1), together with the SOI (6), forms the perturbation
\[
V_{pert}(1, 2) = \sum_{j=1,2} \tilde{V}(x_j, y_j) - V(\vec{r}_1, \vec{r}_2) + \hat{H}_{vdW},
\]
(20)
which is responsible for removing spin degeneracy of states contributing to (19). The eigenvalue problem
\[
(H_0 + V_{pert})|\tilde{\psi}\rangle = E|\tilde{\psi}\rangle,
\]
(21)
where \(H_0\) is the sum of kinetic energy and confinement potential (4), is formulated as a \(4 \times 4\) matrix problem by multiplying (21) by the bra \(\langle s_1 s_2 | \varphi(1, 2)\) from the left (here \(s_{1,2} = \uparrow \) or \(\downarrow\)) and integrating the result over the whole space. The obtained exchange Hamiltonian for the rotated spins \(\tilde{\sigma}\) is of the form (17) with
\[
J = \frac{3}{2} m \omega^2 a^2 e^{-m \omega a^2/2}, D = \frac{32m \alpha^2}{9 \omega a^2 \hat{a}}.
\]
(22)
while \( b = 1, \Gamma = 0 \) to this order. The calculation sketched is valid in the large separation limit, \( a \gg 1/\sqrt{m_3} \), and its most important feature is the scaling \( D \sim a_s^2 \) between the DM coupling and the spin-orbital one. This result is due to the fact that \( O(\alpha_R^2) \) term in \( [23] \) excites both \( x \) and \( y \) oscillators. Since the wave function \( [23] \) contains only the ground states of the oscillators, the \( O(\alpha_R^2) \) term drops out and the first asymmetric correction originates in \( O(\alpha_R^2) \) terms of \([3]\). We checked that this crucial feature is not an artifact of the HL approximation and is also obtained from a more reliable “median-plane” approach \([21, 22, 23]\), which we initiated.

Noting that the DM term \( D \hat{\sigma}_1^{\dagger} \hat{\sigma}_1 \times \hat{\sigma}_2 \) affects the eigenvalue of the two-spin problem only in \( D^2 \) order, we conclude that exchange asymmetry due to the spin-orbit interaction may appear only in \( \alpha_R^2 \) or higher order. This is because the effect of \( \Gamma \)-term in \( [17] \) on the eigenvalues is of first order in \( \Gamma \), and our calculation shows that \( \Gamma \sim O(\alpha_R^2) \). Being proportional to \( J \), see \([22]\), this contribution is also exponentially small. We then conclude that the leading source of spin anisotropy is provided by the vdW contribution \([11]\) and \([13]\), which does not contain an exponential smallness of the exchange.

**Estimate of the vdW coupling.** We now turn our attention to physical manifestations of the vdW spin coupling in the Wigner crystal. Neglecting the exchange interaction for the moment, we consider a two-electron problem within the frozen lattice approximation in which all other electrons are assumed fixed in their equilibrium lattice positions. The potential energy then is just that of four harmonic oscillators \([24]\) with frequencies \( \omega_{x,y} = \sqrt{(\gamma \pm 2)/(m^2 a_B^2 \alpha)} \) and \( \omega_{u,v} = \sqrt{(\gamma \pm 1)/(m^2 a_B^2 \alpha)} \), in notations of Ref \([24]\). Here \( \gamma \approx 5.52 \), \( a_B = \kappa/(m e^2) \) is the Bohr radius, \( \kappa \) is the dielectric constant and \( a \) is the lattice constant of the electron crystal, inversely proportional to the electron density \( n \): \( a = (2/\sqrt{3}n)^{1/2} \).

Repeating the steps that led to \([11]\) we obtain for the Wigner crystal problem

\[
P_{\text{vdW}}^{\text{wigner}} = m^2 a_B^4 \hat{\sigma}_1^{\dagger} \hat{\sigma}_2^{\dagger} \hat{\sigma}_1 \hat{\sigma}_2 = g_{\text{vdW}} \hat{\sigma}_1^{\dagger} \hat{\sigma}_2^{\dagger} \hat{\sigma}_1 \hat{\sigma}_2 \tag{23}
\]

where \( B = [\phi(\omega_x, \omega_u) + \phi(\omega_y, \omega_u) - \phi(\omega_y, \omega_u)]/8 = -3.75 \times 10^{-3} \sqrt{m^2 a_B^2 \alpha} \). The spin-orbit mediated ferromagnetic coupling removes extensive spin degeneracy of the crystal, suppressing the Pomeranchuk effect physics \([4]\). Being of non-frustrated nature, it establishes long-range magnetic order of Ising type with the ordering temperature of the order of the vdW constant \( g_{\text{vdW}} \) \([23]\). It should be compared with the much studied Heisenberg exchange \( J_{\text{exc}} = c(r_s) \exp[-1.612 \sqrt{r_s}], \) expressed in Rydbergs \( R = 1/(2ma_B^2) \). Here \( r_s = 1/\sqrt{\pi a_B^2 n} \) is the dimensionless measure of the interaction strength, and the pre-factor \( c(r_s) \) is a smooth function of it \([3]\). We find that \( g_{\text{vdW}} \) dominates the exchange for \( r_s > r_s^* \approx 20 \) in InAs, which has \( \alpha_R \approx 1.6 \times 10^4 m/s \) \([25]\). For GaAs, with \( \alpha_R \approx 300 m/s \) \([26]\), more diluted situation is required, \( r_s^* \approx 40 \). Given that multi-particle ring-exchange processes on the triangular lattice strongly frustrate any ordering tendencies due to the exchange \([3]\), it appears that our estimate is just a lower bound on the critical density below which spin-orbit-induced ferromagnetic state should be expected.

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