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Bound Electron Pairs Formed by the Spin–Orbit Interaction in 2D Gated Structures

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The bound electron pairs (BEPs) arising due to the pair spin–orbit interaction (PSOI) in 2D structures are explored with a gate that allows the BEPs to be manipulated. The gate breaks the in-plane reflection symmetry of the pair Coulomb field and creates a one-particle Rashba spin–orbit interaction. It is found that the normal component of the electric field substantially affects the BEPs but the key role in forming the BEPs belongs to the in-plane component. The ground state of a BEP with zero total momentum, which is doubly degenerate in the absence of the gate, splits into two states. One of them is tunable by varying the gate voltage, whereas the other is on the contrary robust. The tunable BEP has a higher binding energy which grows as the gate voltage increases, with its orbital and spin structure changing continuously. At large negative voltage, the tunable BEP decays. The orbital and spin structure of the robust BEP does not depend on the gate voltage. Its energy level crosses the conduction band bottom at high gate voltage of any polarity, but the robust BEP remains bound and localized even when in continuum.

The pair interaction of particles depends not only on the their charge and mutual distance but also on their spins and momenta. This well-known fact of the relativistic quantum mechanics[1] is still too poorly studied for the electrons in crystals. However, it becomes important for modern materials with a strong Rashba spin–orbit interaction (SOI).

In the relativistic quantum mechanics, the pair interaction of the electrons moving with small velocity $v/c \ll 1$ is described by the Breit–Pauli Hamiltonian.[1] The pair interaction Hamiltonian derived from it in the frame of the $k \cdot p$ approximation[2] has a form very similar to the original Breit–Pauli Hamiltonian with an important difference that the material-dependent coefficients appear in each of its terms.

Of most interest is the SOI component of the pair interaction because it couples the spin and orbital degrees of freedom, which can essentially affect the dynamics of interacting electrons and result in new collective states. The pair SOI (PSOI) produced by the Coulomb fields $\mathbf{E}(\mathbf{r}_i - \mathbf{r}_j)$ of interacting electrons has the following form[3]

$$H_{\mathrm{PSOI}} = \frac{\alpha}{\hbar}\sum_{i\neq j}(\mathbf{p}_i \times \mathbf{E}(\mathbf{r}_i - \mathbf{r}_j)) \cdot \boldsymbol{\sigma}_i \qquad (1)$$

where $\mathbf{p}_i$ is the momentum of the ith electron, $\boldsymbol{\sigma}$ is the Pauli vector, and α is a material-dependent SOI constant. Having been calculated within $k \cdot p$ approximation, when the Coulomb field is assumed to be a smooth function on the scale of the lattice constant, the value of α is the same as the Rashba constant of the material.

The most interesting feature of the PSOI is that it creates attraction between the electrons in certain spin configurations tied to their momenta. A completely unusual property of such an attraction is the fact that it is determined directly by the electric field and, therefore, for the Coulomb interaction, it is especially large at small distances between the particles and rapidly decreases at large distances. Thus, the pair interaction we consider here is attractive on a small scale and repulsive on the large distance. The bound electron pairs (BEPs) formed as a result of this interaction[4–6] are drastically different from other composite particles, which are currently widely studied in bulk materials,[7,8] low-dimensional systems,[9–13] and even for cold atoms in optical lattice.[14]

Another feature of the PSOI is that it depends on the configuration of the Coulomb field which acts between electrons and can be controlled by a gate in low-dimensional systems. To find out how the field configuration affects BEPs and, in particular, their binding energy characterizing the stability of the pairs, we focus here on considering the two-body problem, which can be solved exactly. Considering the possible implementations, the study of isolated pairs is of interest for low-dimensional structures, such as quantum dots and quantum cavities. The properties of isolated pairs are also worth studying for 2D materials with not very high Fermi energy, when the Fermi wave vector is small on the scale of the reciprocal value of the region size, where the pair attraction prevails. In this case, the electrons near and below the Fermi energy as well as the pairs strongly repel each other; therefore, the Cooper instability does not occur.

In a 2D system symmetric at the in-plane reflection, PSOI is created by the electric field acting in the plane of the system.[5,6] On the contrary, in 1D systems, PSOI is created only by a normal component of the field, which arises when the axial symmetry of

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the system is broken, for example, by a proximate gate. In this case, PSOI originates from image charges induced by the interacting electrons at the gate[15] and also leads to BEP formation.[4]

The spectrum of the BEPs together with their spin structure is quite different, depending on whether the PSOI is produced solely by the in-plane field or by the normal electric field. Despite the difference in the electric field configuration in both cases, one can still classify the BEPs according to the nature of the electron motion, which produces the PSOI, to arrive at two distinct types of BEPs.

The relative motion of electrons in the pair creates the *relative* BEPs. In the symmetric 2D case, the relative BEPs are triplet-like states with parallel spins, with the ground state of the BEP being doubly degenerate. In contrast, the relative BEPs in the 1D gated wire are of a mixed singlet–triplet type.

The motion of an electron pair as a whole forms the *convective* BEPs. Their binding energy crucially depends on the total momentum of the pair and the spin structure is more complicated. In symmetric 2D systems, the convective BEPs do not possess a definite spin. In contrast, in the gated 1D wire, the spin projection is well defined so that $S_z = \pm 1$, its sign being determined by the direction of the center-of-mass momentum.

Of great interest is the problem of BEPs in 2D systems with a metal gate, because one can anticipate a unique opportunity to control their binding energy and the spin state. This raises the question of how the properties of BEPs change in such structures. The main effects are because the symmetry of the Coulomb fields inherent in the two cases mentioned previously is broken in such structures. The electric field has both in-plane and normal components, the interplay of which creates a nontrivial configuration of the effective magnetic field acting on the electron spin. The BEP spin state is not predefined, but rather should be determined self-consistently, together with its orbital structure via the quantum-mechanical equations of motion considering the particular field configuration.

Due to the symmetry breaking of the electric fields, it is no longer possible to separate the relative motion of the particles from the motion of the center of mass; therefore, the relative and convective states are mixed. In this article, we solve this intricate problem in the case when BEP has a zero total momentum without any restrictions on the relative magnitude of the tangential and normal components of the electric field. As a result, we came to the conclusion that the in-plane component plays a key role in the formation of BEPs, and the presence of a normal component leads to the radical reconstruction of BEPs.

Specific calculations are carried out for a model system that consists of an atomically thin layer of material with a strong Rashba SOI separated by a spacer from a charged metallic gate, as shown in **Figure 1**. The presence of the gate affects both the spatial configuration and the magnitude of the Coulomb field of the interacting electrons. In addition, the external voltage applied to the gate creates one-particle Rashba SOI. We explore how these factors govern BEP formation, their spectrum, and spin structure.

The normal field lifts the degeneracy of the relative BEPs to produce two kinds of BEPs having very different properties. Our most interesting finding is that there appears a *robust* BEP that remains unchanged with the variation of gate potential. At a large enough gate voltage of any sign, the robust BEP gets into the continuum of the band states, where it remains

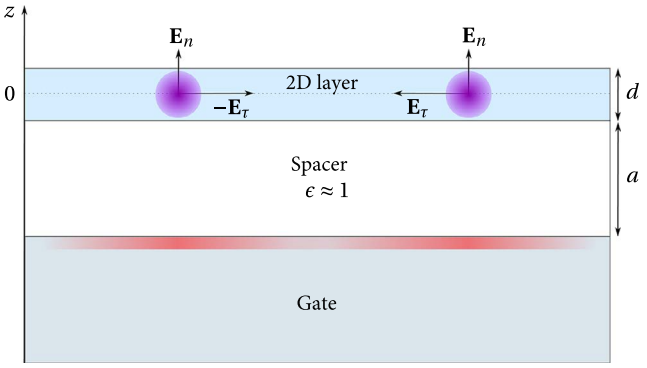

Figure 1. A 2D layer separated from a metallic gate by a spacer made of a weak dielectric. Normal ($\mathbf{E}_n$) and in-plane ($\mathbf{E}_\tau$) components of the electric field acting on each electron are created by a neighboring electron and the polarization charges, as well as the total charge of the gate.

localized. On the contrary, the BEP of the other kind is tunable by the gate voltage. The positive voltage applied to the gate increases its binding energy, whereas the negative voltage moves the energy level of the *tunable* BEP to the continuum, where it decays.

Consider two electrons at positions $\mathbf{r}_i$ in the 2D system. In the two-particle basis $\{|\uparrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\downarrow\downarrow\rangle\}$, the system wave function represents a Pauli spinor of the fourth rank, $\Psi(\mathbf{r}_1, \mathbf{r}_2) = (\Psi_{\uparrow\uparrow}, \Psi_{\uparrow\downarrow}, \Psi_{\downarrow\uparrow}, \Psi_{\downarrow\downarrow})^\intercal$. Introduce the relative electron position $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$, the center-of-mass position $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$, and the corresponding momenta $\mathbf{p} = -i\hbar\nabla_\mathbf{r}$ and $\mathbf{P} = -i\hbar\nabla_\mathbf{R}$. The PSOI Hamiltonian, built as the Kronecker sum of the terms in Equation (1), is equal to

$$H_{\mathrm{PSOI}} = \frac{\alpha}{2\hbar}\begin{pmatrix} \frac{4E_\tau(r)}{r}(\mathbf{r}\times\mathbf{p})_z & -\xi_+ + \Xi_+ & \xi_+ + \Xi_+ & 0 \\ -\xi_- + \Xi_- & \frac{2E_\tau(r)}{r}(\mathbf{r}\times\mathbf{P})_z & 0 & \xi_+ + \Xi_+ \\ \xi_- + \Xi_- & 0 & -\frac{2E_\tau(r)}{r}(\mathbf{r}\times\mathbf{P})_z & -\xi_+ + \Xi_+ \\ 0 & \xi_- + \Xi_- & -\xi_- + \Xi_- & -\frac{4E_\tau(r)}{r}(\mathbf{r}\times\mathbf{p})_z \end{pmatrix} \quad (2)$$

with $\xi_\pm = [F(r), \gamma_\pm]_+$ and $\Xi_\pm = F(r)\Gamma_\pm$. The normal field $F(r) \equiv E_n(r)$ for the particular geometry considered in Figure 1 is given by Equation (7), Supporting Information. Then, $\Gamma_\pm = P_y \pm iP_x$ and

$$\gamma_\pm = p_y \pm ip_x = \hbar e^{\mp i\phi}\left(\pm\partial_r - \frac{i}{r}\partial_\phi\right) \quad (3)$$

The anticommutator $[\hat{A}, \hat{B}]_+ = \hat{A}\hat{B} + \hat{B}\hat{A}$ is introduced to maintain the hermiticity of the Hamiltonian while projecting Equation (1) to the 2D subspace.

A single-particle SOI contribution can be included in Equation (2) by adding the field

$$F_g = 4\pi n_g + E_n(0) \quad (4)$$

produced by the gate surface charge density n_g and the electron's own image, to the normal field $F(r)$, so that the total normal field becomes $F(r) = E_n(r) + F_g$.

Later, we restrict ourselves to a particular case of $P = 0$, when the BEPs are essentially the relative ones.

The equation of motion for the two-body wave function follows from the full Hamiltonian

$$H = H_{\mathrm{PSOI}} + V + T \tag{5}$$

which, in addition to the PSOI of Equation (2), contains diagonal contributions coming from the electron–electron (e–e) repulsion $V(r)$ of Equation (6), Supporting Information, and the kinetic energy T. In what follows, it is convenient to introduce the shift $eF_g a$ in the energy and the potential V. This eliminates the trivial effect of gate potential and allows us to consider only the effect of the normal electric field. For simplicity, we consider here a minimal model with quadratic band dispersion.

In the absence of the gate, when $F(r) \equiv 0$, the relative BEPs represent degenerate pairs of triplet states with the spin orientation tied to the angular momentum direction.[5,6] The lowest-lying states, corresponding to the minimum possible angular momentum $l = \pm 1$, are

$$\Psi_{-}(\mathbf{r}) = (u(r)e^{-i\phi}, 0, 0, 0)^{\intercal} \tag{6}$$

and

$$\Psi_{+}(\mathbf{r}) = (0, 0, 0, u(r)e^{i\phi})^{\intercal} \tag{7}$$

The radial wave function $u(r)$ is determined from the Schrödinger equation

$$\left[T_1 + V(r) - 2\alpha\frac{E_\tau(r)}{r}\right]u(r) = \varepsilon_0 u(r) \tag{8}$$

where T_l stands for kinetic energy including the centrifugal potential

$$T_l = -\frac{\hbar^2}{m}\left(\frac{d^2}{dr^2} + \frac{1}{r}\frac{d}{dr} - \frac{l^2}{r^2}\right), \quad l = 0, \pm 1, \ldots \tag{9}$$

The binding potential produced by PSOI is the last term on the left hand side of Equation (8). Considering the short-range asymptotics of the in-plane field E_τ given by Equation (9), Supporting Information, we see that the BEPs are formed by the singular attractive potential $\propto -\frac{\alpha}{\chi r^2}$, with χ being the 2D susceptibility of the layer. This overcomes the centrifugal barrier for a sufficiently large α, let alone a much weaker Rytova–Keldysh repulsion $\propto \log\frac{r}{2\pi\chi}$.

The $-1/r^2$ potential leads to a fall to the center,[16] unless a short-range cut-off is introduced. The regularization of the binding potential can be caused by mechanisms such as the Zitterbewegung of electrons in crystalline solids or natural cutting-off due to averaging the 3D quantities across the layer thickness. We regularize the potential by imposing a zero boundary condition for the wave function at the cut-off length of the order of the layer thickness.[5,6] Then binding energy can be estimated as

$$|\tilde{\varepsilon}_0| = \frac{x_1^2(\lambda)}{(d/a_B)^2} \tag{10}$$

where $x_1(\lambda)$ is the first (largest) zero of the Macdonald function $K_{i\lambda}(x)$, and the amplitude of the attraction is defined as

$$\lambda = \sqrt{\frac{4\tilde{\alpha}}{d/a_B} - 1} \tag{11}$$

Here, we introduced a convenient dimensionless SOI constant $\tilde{\alpha} = \alpha/ea_B^2$, with the Bohr radius in the material $a_B = \epsilon\hbar^2/me^2$. The BEPs appear as soon as $\tilde{\alpha} > d/4a_B$, which is attainable in materials such as Bi_2Se_3,[17] BiTeI,[18,19] or BiSb monolayers,[20] for which $\tilde{\alpha}$ is of the order of unity.[21] From now on, the energy with a tilde is given in 2Ry units, with the Rydberg constant in the material being $\mathrm{Ry} = \hbar^2/2ma_B^2$. Equation (10) gives $|\varepsilon_0|$ on the level of tens of Rydberg.

The normal field $F(r)$ that appears in the presence of the gate lifts degeneracy. In the lowest order of degenerate perturbation theory, a perturbation does so by mixing the states with certain weights defined by its matrix elements.[16] However, the corresponding matrix elements calculated with the states of Equation (6) and (7) are all zero. Consequently, a higher-order approximation should be used which, generally speaking, involves the scattering states of the Hamiltonian of Equation (5) in perturbation expansion. Fortunately, this tedious procedure can be avoided by checking that the state

$$\Psi(\mathbf{r}) = (u(r)e^{-i\phi}, 0, 0, -u(r)e^{i\phi})^{\intercal} \tag{12}$$

where $u(r)$ given by Equation (8) provides the exact solution of the full Hamiltonian (5) for the arbitrary magnitude of the normal field $F(r)$. This antisymmetric combination of the unperturbed solutions of Equation (6) and (7) obviously does not include any scattering states, which would depend on the normal field $F(r)$.

It follows from Equation (8) that neither the radial wave function $u(r)$, nor the energy ε_0 depends on the normal field $F(r)$. Therefore, the bound state of Equation (12) is robust with its orbital and spin structure unaffected by the normal electric field applied to the system. The result is not specific to a particular sandwich geometry considered here and holds for any profile of $F(r)$ provided that 1) there is no external field parallel to the layer and 2) the electron pair has a zero total momentum.

Note that the binding energy is measured from the bottom of the conduction band ε_c, which in the presence of the SOI is shifted as soon as $F_g \neq 0$. For a pair of electrons, its position is given by

$$\tilde{\varepsilon}_c = -\frac{1}{4}\mathfrak{F}_g^2 \tag{13}$$

where $\mathfrak{F}_g$ stands for the gate field F_g of Equation (4) normalized according to $\mathfrak{F}_g = \tilde{\alpha}F_g/F_0$, with $F_0 = e/2\epsilon a_B^2$. In 1D quantum wires, the BEPs always lie below ε_c.[4] This is not, generally speaking, the case in a 2D system. Increasing F_g lowers ε_c, keeping ε_0 intact, so eventually the energy level ε_0 gets into the conduction band. According to Equation (8), the robust BEP remains localized even at $\varepsilon_0 > \varepsilon_c$. In other words, there appears a discrete energy level in the continuum that does not mix with the band states.

Contrary to Equation (12), the symmetric combination of Equation (6) and (7) is not a solution at $F \neq 0$. All four spinor components do arise in the exact solution, which reads as

$$\Psi(\mathbf{r}) = (u(r)e^{-i\phi}, v(r), -v(r), u(r)e^{i\phi})^{\mathrm{T}} \quad (14)$$

This form ensures the antisymmetry of Ψ with respect to the permutation of electrons. From the point of view of the perturbation theory, Equation (14) includes the contribution from the scattering states of Equation (5), which makes it sensitive to the normal field $F(r)$.

The radial wave functions satisfy the system of equations

$$\begin{cases} \left[T_1 + V(r) - 2\alpha\frac{E_c(r)}{r}\right]u - \alpha\left[2F(r)\frac{d}{dr} + F'(r)\right]v = \varepsilon u \\ [T_0 + V(r)]v + \alpha\left[2F(r)\left(\frac{1}{r} + \frac{d}{dr}\right) + F'(r)\right]u = \varepsilon v \end{cases} \quad (15)$$

which should be solved with zero boundary conditions at $r = d$ and at infinity. We are mostly interested in the dependence of the binding energy on the normal electric field.

The analytical treatment of this problem is expounded in Supporting Information. Here, we solve Equation (15) numerically with the exact interaction potential and field of Equation (6) and (7), Supporting Information. To give an estimate of the binding energy of the tunable state, consider the system based on Bi_2Se_3, for which $\alpha \approx 1300\,\mathrm{e\,\AA^2}$,[21] $a_B \approx 52\,\text{Å}$, and hence $\tilde{\alpha} \approx 0.47$. For a reasonable value of the electric field $F_g = 10^5\,\mathrm{V\,cm^{-1}}$, the layer thickness of $d = 28.7\,\text{Å}$, corresponding to three quintuple layers of Bi_2Se_3, and the distance to the gate $a = 2d$, we obtain $|\varepsilon| = 40\,\mathrm{meV}$.

In **Figure 2**, we plot the energy levels of the robust BEP of Equation (12) and the tunable BEP of Equation (14) versus the normalized field of the gate for the model system with $\tilde{\alpha} = 1$, $\chi = 0.4a_B$, $d = 0.25a_B$, and $a = a_B$. In addition, the position of the bottom of the conduction band is shown. At a large negative voltage applied to the gate, the tunable BEP gets into the continuum where it decays, whereas the positive voltage facilitates the pairing by increasing the binding energy. The binding energy of the robust BEP, measured from ε_c, decreases when the voltage is applied to the gate, so at a large gate voltage of any polarity, the energy level crosses the continuum boundary, but the robust BEP remains bound and localized even in the continuum.

We studied the BEPs formed by the PSOI in the most realistic and practically important situation of a 2D system with a gate,

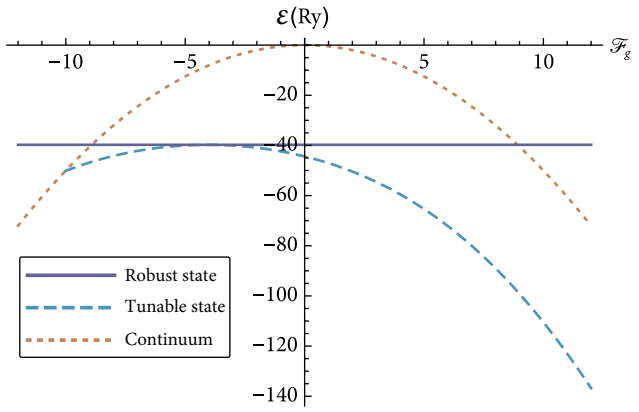

Figure 2. The dependence of the binding energy of the BEPs, as well as the bottom of the conduction band, on the normalized gate field.

when the PSOI is created by a Coulomb field of interacting electrons, having both in-plane and normal components. We focus on the effects due to the interplay of these components for a particular case of a BEP with zero total momentum.

We have found that the normal field lifts the degeneracy of the pair of bound states created by the in-plane field. One of the resulting BEPs that has a higher binding energy is tunable by gate voltage. Its binding energy is significantly increased by the positive gate voltage, and its spin and orbital structure continuously transform while changing $\mathfrak{F}_g$.

In contrast, the second state demonstrates a totally unexpected behavior. Its spin and orbital structure does not depend on the gate voltage, and its energy varies exactly as the potential induced by the gate at the layer. The binding energy measured from the conduction band bottom decreases with the gate voltage, so at a large $\mathfrak{F}_g$ the energy level gets into the continuum of the band states. It is interesting that this state remains bound and localized even when it is in the continuum. The fact that the BEP is so stable with respect to the normal electric field evidences that the in-plane electric field of the Coulomb interaction plays a key role in the electron pairing in competition with the normal component.

The behavior of electrons in the presence of the PSOI in a many-electron system needs further serious study. One can expect that because of the unusual form of the pair interaction, various scenarios are possible, such as the formation of electronic complexes, spontaneous symmetry breaking, and of course the formation of superconducting phases.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

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