Unconventional Pairing in Excitonic Condensates under Spin-Orbit Coupling

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It is shown that Rashba and Dresselhaus spin-orbit couplings enhance the conclusive power in the experiments on the excitonic condensate by at least three low temperature effects. First, spin-orbit coupling facilitates the photoluminescence measurements via enhancing the bright contribution in the otherwise dominantly dark ground state. The second is the presence of a low temperature power law dependence of the specific heat and weakening of the second order transition at the critical temperature. The third is the appearance of the nondiagonal elements in the static spin susceptibility.

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The existence of the excitonic collective state in bulk semiconductors was speculated in the early 1960s [1]. The idea was that, due to the attractive interaction between the electron-hole pairs, excitons act like single bosons in low densities, and consequently, they should condense in 3D at sufficiently low temperatures. The difference of the speculated excitonic Bose-Einstein condensate (BEC) from the atomic BEC is that, due to the small exciton band mass ($m_x \simeq 0.07 m_e$ where m_e is the bare electron mass), the condensation should occur at much higher critical temperatures than the atomic BEC. One of the first experiments was on bulk CuO_2 samples [2,3]. In the last 15 years, however, experimental efforts were primarily focused on coupled quantum wells (CQW). The motivation is that, spatially indirect excitons have much longer lifetimes (in the order of 10 μs) in the presence of an additional electric field applied perpendicular to the plane, in contrast with the bulk where lifetimes are on the order of ns. Recent reviews [4] with CQWs remark the evidence of the exciton condensate (EC) in the measurements of the large indirect exciton mobility and radiative decay rates, enhancements in exciton scattering rate, and the narrowing of the PL spectra. It is also suggested that there is room for different conclusions other than the excitonic BEC [4,5].

Here, we propose an alternative method to examine the EC by including the spin degree of freedom of the electrons (e) and holes (h) via spin-orbit coupling (SOC) of the Rashba [6] (RSOC) and the Dresselhaus [7] (DSOC) types. The RSOC is manipulated by the external electric (E) field whereas the DSOC is known to be intrinsic in zinc-blende structures [8]. Major differences exist between the SOC induced effects in the noncentrosymmetric superconductors and the semiconductor electron-hole (e-h) CQWs. In the former, the SOC is stronger than the typical condensation energy, i.e., $\Delta_0 \lesssim E_{so} \ll E_F$ where E_{so} and Δ_0 are typical SOC and condensation energies [9] and E_F is the Fermi energy, whereas in the latter $E_{so} < \Delta_0 \ll E_F$ [10]. It may thus seem that such a perturbative effect may not play a significant role in the thermodynamics of the latter case. However, manipulating the spin changes the nature of the ground state as well as the low temperature behavior of the thermodynamic observables. Three distinct effects of the SOC are expected in the EC as (i) a controllable mixture of the dark and bright condensates (DC and BC hereon) in the ground state, (ii) finite off diagonal static spin susceptibilities, and (iii) nonisotropic narrowing between the two lowest energy bands near E_F manifesting at low temperatures.

The reasons for the lack of conclusive evidence for the EC in the PL measurements was recently suggested in relation to (i) above [11]. It was proposed that the Pauli exclusion principle should implement a ground state predominantly composed of DC which does not couple to light due to its spin (± 2). It is claimed that it is mainly the BC [spin (± 1)] that is probed in the PL experiments which is energetically above the DC by 0.1 μeV. Thus, the Pauli principle yields a weak splitting in the effective interband Coulomb strengths between the dark and the bright channels breaking the spin rotation symmetry within the individual layers. Here, we take the relevant fundamental symmetries [i.e., spin degeneracy (S), reflection symmetry (P), rotational invariance (R) of the Coulomb interaction, time reversal symmetry (T), and the fermion exchange symmetry (FX)] into full account. It is confirmed that the ground state is dominated by the DC in the absence of the SOC. In most semiconductors, the SOC is in the range 0.1–10 meV for RSOC and 1–30 meV for the DSOC with the larger ones being for the indium based materials at typical densities $n_x \lesssim 10^{11}$ cm^{-2}. We demonstrate that a weak SOC can change the ground state from dominantly DC to a controllable mixture of DC and BC [12]. It is crucial that such observations are expected to be more conclusive than detecting the DC indirectly by its influence on the line shift of the BC [11]. Additionally, and in relation to (ii), the SOC permits the off diagonal spin susceptibility to be finite, enabling complementary measurements. In relation to (iii), there are additional effects of the SOC on the band structure reminiscent of the zero temperature lines-of-nodes observed in the superconducting gap of the noncentrosymmetric superconductors.

There, due to the anisotropy of the strong SOC, the zero temperature superconducting gap vanishes at certain lines on the Fermi surface changing the temperature behavior of the thermodynamic quantities from exponentially suppressed to power law [13]. In the present case, a similar effect at low temperatures is produced in the presence of the SOC.

We demonstrate these three effects (i–iii) by considering a model of e-h type CQWs confined in the x-y plane with z [(0,0,1) of the underlying lattice] as the growth direction. The CQWs are separated by a tunneling barrier of thickness $d \simeq 100$ Å, and the width of each well is $W = 70$ Å. The model Hamiltonian is

$$\mathcal{H}_X = \sum_{\vec{k},\sigma,p} (\zeta^{(p)}_{\vec{k}} - \mu_p)\hat{p}^\dagger_{\vec{k},\sigma}\hat{p}_{\vec{k},\sigma} + \mathcal{H}^{(e)}_{\rm so} + \mathcal{H}^{(h)}_{\rm so} + \frac{1}{2}\sum_{\substack{\vec{k},\vec{q}\\ \sigma,\sigma',p,p'}} V_{pp'}(q)\hat{p}^\dagger_{\vec{k}+\vec{q},\sigma}\hat{p}'^\dagger_{\vec{k}'-\vec{q},\sigma'}\hat{p}'_{\vec{k}',\sigma'}\hat{p}_{\vec{k},\sigma} \tag{1}$$

where μ_p is the chemical potential for electrons ($p = e$) and heavy holes ($p = h$). Here, $\hat{p}_{\vec{k}\uparrow}$ indicates the annihilation operator at the wave vector $\vec{k}$ and spin $= +1/2$ for $p = e$, and spin $= +3/2$ for $p = h$. The spin independent *bare* single particle energies are $\zeta^{(p)}_{\vec{k}} = \hbar k^2/(2m^*_p)$ where m^*_p is the effective mass ($m^*_e = 0.067 m_e$ and $m^*_h = 0.4 m_e$ where m_e is the free electron mass) and $V_{pp'}(q) = s_{pp'} 2\pi e^2/(\epsilon_\infty q)$ is the Coulomb interaction between p and p' particles where $s_{ee} = s_{hh} = 1$ and $s_{eh} = -2$. The CQWs are formed within the GaAs-AlGaAs interface. The electron R&D SOCs are described by $\mathcal{H}^{(e)}_{\rm so}$ as [in the basis $(\hat{e}_{\vec{k}\uparrow}\hat{e}_{\vec{k}\downarrow})$],

$$\mathcal{H}^{(e)}_{\rm so}(\vec{k}) = \begin{pmatrix} 0 & i\alpha^{(e)}_R k_- + \alpha^{(e)}_D k_+ \\ -i\alpha^{(e)}_R k_+ + \alpha^{(e)}_D k_- & 0 \end{pmatrix} \tag{2}$$

where $\alpha^{(e)}_R = r^{6c6c}_{41} E_0$ and $\alpha^{(e)}_D = -b^{6c6c}_{41}\langle k_z^2\rangle$ with $\vec{E} = E_0\vec{e}_z$ as the external E field, and $r^{6c6c}_{41} \simeq 5.2e$ Å^4, $b^{6c6c}_{41} \simeq 27.6$ eV Å^3 are the coupling constants for GaAs [8] with $\langle k_z^2\rangle \simeq (\pi/W)^2$. The hole R&D SOCs are described by $\mathcal{H}^{(h)}_{\rm so}$ as [in the basis $(\hat{h}_{\vec{k}\uparrow}\hat{h}_{\vec{k}\downarrow})$],

$$\mathcal{H}^{(h)}_{\rm so}(\vec{k}) = \begin{pmatrix} 0 & \alpha^{(h)}_R k^3_- + \alpha^{(h)}_D k_+ \\ \alpha^{(h)}_R k^3_+ + \alpha^{(h)}_D k_- & 0 \end{pmatrix} \tag{3}$$

where $\alpha^{(h)}_R = \beta_h E_0$ and $\alpha^{(h)}_D = -b^{8v8v}_{41}\langle k_z^2\rangle$ with $\beta_h \simeq 7.5 \times 10^6 e$ Å^4, $b^{8v8v}_{41} \simeq -81.9$ eV Å^3 as the coupling constants for the holes [8]. In the e-h basis $(\hat{e}_{\vec{k}\uparrow}\hat{e}_{\vec{k}\downarrow}\hat{h}^\dagger_{-\vec{k}\uparrow}\hat{h}^\dagger_{-\vec{k}\downarrow})$, the condensed system has the reduced 4×4 Hamiltonian (up to a multiple of the unit matrix)

$$\mathcal{H}_X = \begin{pmatrix} \mathbf{\Sigma}^{(e)}(\vec{k}) - \mu_x & \mathbf{\Delta}^\dagger(\vec{k}) \\ \mathbf{\Delta}(\vec{k}) & \mathbf{\Sigma}^{(h)}(-\vec{k}) + \mu_x \end{pmatrix} \tag{4}$$

where μ_x is the exciton chemical potential found self-consistently by conserving the number of exciton pairs [10,14] and the diagonals are the e and h single particle energies

$$\mathbf{\Sigma}^{\binom{e}{h}}(\vec{k}) = \begin{pmatrix} \pm\tilde{\zeta}_{\vec{k}\uparrow} & \pm\Sigma^{\binom{e}{h}*}_{\uparrow\downarrow}(\vec{k}) + \mathcal{H}^{\binom{e}{h}}_{\rm so}(\vec{k}) \\ \pm\Sigma^{\binom{e}{h}}_{\uparrow\downarrow}(\vec{k}) + \mathcal{H}^{\binom{e}{h}*}_{\rm so}(\vec{k}) & \pm\tilde{\zeta}_{\vec{k}\downarrow} \end{pmatrix}. \tag{5}$$

where $\tilde{\zeta}_{\vec{k}\sigma} = [\tilde{\zeta}^{(e)}_{\vec{k}\sigma} + \tilde{\zeta}^{(h)}_{\vec{k}\sigma}]/2$ includes the diagonal self energies, i.e., $\tilde{\zeta}^{(p)}_{\vec{k}\sigma} = \zeta^{(p)}_{\vec{k}} + \Sigma^{(p)}_{\sigma\sigma}(\vec{k})$. The off diagonal components $\Sigma^{(p)}_{\uparrow\downarrow} = \Sigma^{(p)*}_{\downarrow\uparrow}$ in Eq. (5) represent the self energies of cross spin correlations. The nondiagonal elements in Eq. (4) are the spin dependent excitonic order parameters

$$\mathbf{\Delta}(\vec{k}) = \begin{pmatrix} \Delta_{\uparrow\uparrow}(\vec{k}) & \Delta_{\downarrow\uparrow}(\vec{k}) \\ \Delta_{\uparrow\downarrow}(\vec{k}) & \Delta_{\downarrow\downarrow}(\vec{k}) \end{pmatrix} \tag{6}$$

with the diagonals corresponding to the DC [11] and the off diagonals corresponding to the BC in the mixture of *triplet* [15] (T) and the *singlet* (S) state $\Delta_{\binom{T}{S}}(\vec{k}) = [\Delta_{\uparrow\downarrow}(\vec{k}) \pm \Delta_{\downarrow\uparrow}(\vec{k})]/2$.

The solution of Eq. (4) includes the self-consistent mean-field Hartree-Fock calculation of the thermodynamic state where

$$\begin{aligned} \Sigma^{(p)}_{\sigma\sigma'}(\vec{k}) &= -\sum_{\vec{q}} V_{pp}(\vec{q})\langle\hat{p}^\dagger_{\vec{k}+\vec{q}\sigma}\hat{p}_{\vec{k}+\vec{q}\sigma'}\rangle, \quad \hat{p} = (\hat{e},\hat{h}) \\ \Delta_{\sigma\sigma'}(\vec{k}) &= \sum_{\vec{q}} V_{eh}(\vec{q})\langle\hat{e}^\dagger_{\vec{k}+\vec{q},\sigma}\hat{h}^\dagger_{-\vec{k}-\vec{q},\sigma'}\rangle \end{aligned} \tag{7}$$

with $\langle\ldots\rangle$ as the thermal average. The symmetries affecting the solution of Eq. (4) are R, T, S, and P. As a fundamental difference of the EC in CQWs from the noncentrosymmetric superconductors, the fermion exchange (FX) symmetry is absent in the former manifesting in the appearance of the "triplet" states even in the presence of T symmetry and the order parameters with a fixed total spin have mixed parities [16]. The transformations corresponding to these symmetries are shown in Table I. In the absence of SOC, the respected symmetries are R, T, S, P, whereas with finite SOC only T is manifest. Application of Table I without the SOC and FX shows that $\Sigma^{(p)}_{\uparrow\downarrow}(\vec{k}) = \Sigma^{(p)^*}_{\downarrow\uparrow}(\vec{k}) = 0$, $\Sigma^{(p)}_{\uparrow\uparrow}(\vec{k}) = \Sigma^{(p)}_{\downarrow\downarrow}(\vec{k}) \neq 0$, and $\Delta_{\uparrow\uparrow}(\vec{k}) = \Delta_{\downarrow\downarrow}(\vec{k}) \neq 0$ for the DC whereas

TABLE I. The fundamental symmetry operations. Note that FX is inapplicable in this work. Here, σ_x, σ_y are the Pauli-Dirac matrices and $\mathbf{X} = \mathbf{\Sigma}^{(p)}$, $\mathbf{\Delta}$ as given by Eqs. (5) and (6).

Symmetry	Action
T	$\mathbf{X}(\vec{k}) \to (-i\sigma_y)\mathbf{X}^*(-\vec{k})(-i\sigma_y)^{-1}$
S	$\sigma_x\mathbf{\Delta}(\vec{k})\sigma_x$
P	$\mathbf{X}(\vec{k}) \to \mathbf{X}(-\vec{k})$
FX	$\mathbf{\Delta}(\vec{k}) \to -\mathbf{\Delta}^T(-\vec{k})$, $\mathbf{\Sigma}^{(e)}(\vec{k}) \leftrightarrow \mathbf{\Sigma}^{(h)}(\vec{k})$

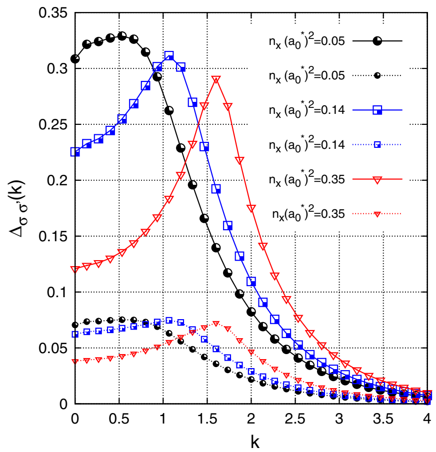

FIG. 1 (color online). The zero temperature order parameters for DC (i.e., $\Delta_{\uparrow\uparrow} = \Delta_{\downarrow\downarrow}$), and BC (i.e., $\Delta_{\uparrow\downarrow}$) without SOC at three densities ranging from BEC [i.e., $n_x(a_0^*)^2 < 0.1$] to BCS [i.e., $0.1 < n_x(a_0^*)^2$] type. Solid and dashed lines, respectively, correspond to DC and BC at the corresponding exciton density.

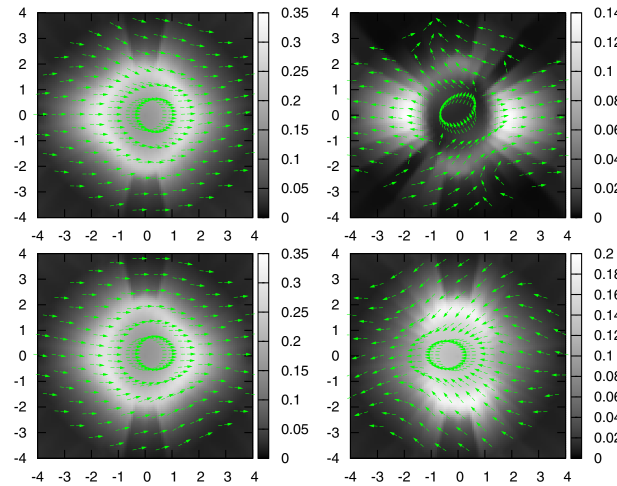

FIG. 2 (color online). The DC and BC as function of $\vec{k}$ with the same parameters used in Fig. 1 but in the presence of R&D SOC. Starting from the top left in counterclockwise, $\Delta_{\uparrow\uparrow}$, $\Delta_{\downarrow\downarrow}$, Δ_S, Δ_T. Gray scales on the right are the magnitudes (in units of E_H), and arrows depict the phase. The SOC parameters used are as given below Eq. (2) and (3).

the BC in the triplet state vanishes, i.e., $\Delta_T(\vec{k}) = 0$ therefore $\Delta_S(\vec{k}) = \Delta_{\uparrow\downarrow}(\vec{k})$. In our numerical calculations, we also break the spin rotation symmetry of the individual layers in Eq. (1) by implementing a weak dark-bright splitting (≈ 0.1 μeV) in the Coulomb strengths in order to account for the Pauli principle. The radial configurations of the isotropic DC and BC order parameters are depicted in Fig. 1 indicating that the bright "singlet" is much weaker than the dark "triplets" and the ground state is dominated by the DC. In all figures, $\vec{k}$ is in units of the exciton Bohr radius a_0^* and all energies are in units of the exciton Hartree energy E_H.

A more interesting case is when the R&D SOCs are present. With only the T symmetry remaining, Eq. (6) is complex with unconventional phase texture particularly in the cross spin configurations. The FX breaking is even stronger due to the different couplings in the conduction and the valence bands. Figure 2 depicts the dark and bright components of Eq. (6) in the presence of R&D SOCs for GaAs with $E_0 = 75$ kV/cm and $n_x(a_0^*)^2 = 0.35$. It is observed that Δ_T and Δ_S have comparable magnitudes to those of the DC.

We have thus far shown that the SOC enhances the BC in the ground state in the context of the first effect (i) above. We now examine the second and the third effects (ii) and (iii) together in the temperature and SOC dependence of the specific heat (C_v) and the nondiagonal spin susceptibility (χ_{xz}). In the absence of SOC, the temperature dependence of C_v is exponentially suppressed for $T \simeq 0$. The critical temperature T_c is identified by the anomaly ΔC_v where the condensate has a second order transition into the e-h liquid. Increasing the SOC decreases T_c [Fig. 3(a)], weakens the anomaly at T_c [Fig. 3(b)], and changes the behavior to a power law near $T \simeq 0$ [as shown for $n_x(a_0^*)^2 = 0.29$ in Fig. 3(c)]. At low densities where the condensation is stronger, the exponential suppression is more robust to changes into a power law than in the case of high densities. The critical temperature T_c is weakly density dependent whereas ΔC_v is much stronger at low densities [Figs. 3(a) and 3(b)]. The overall effect is that

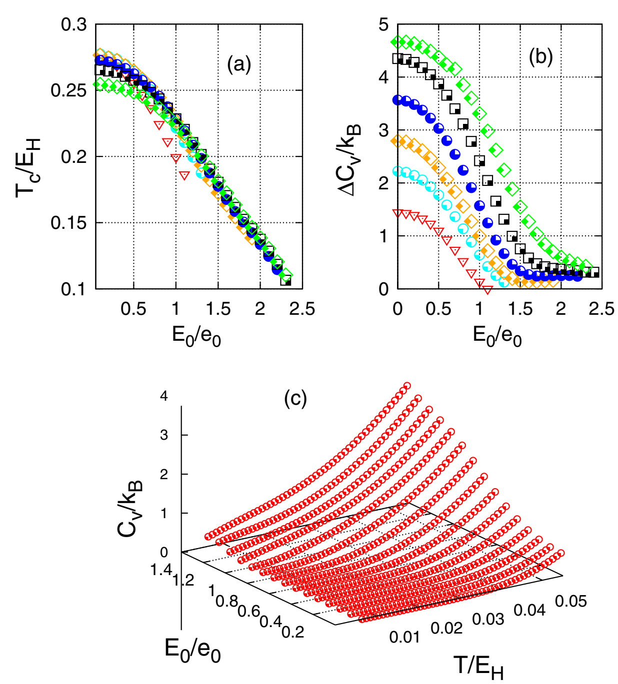

FIG. 3 (color online). (a) T_c/E_H vs E_0, (b) ΔC_v vs E_0, and (c) C_v vs T and E_0 at $T \ll T_c$ for $n_x(a_0^*)^2 = 0.29$. Here, $e_0 = 150$ kV/cm, k_B is the Boltzmann constant. Symbol coding at different n_x is shared between (a) and (b). From the top to bottom plots in (b), the densities are $n_x(a_0^*)^2 = 0.29$, 0.33, 0.38, 0.42, 0.46, 0.62.

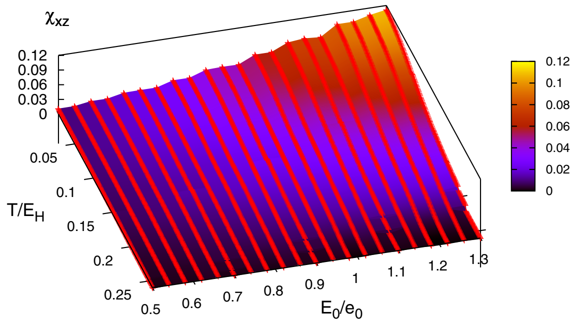

FIG. 4 (color online). The nondiagonal component χ_{xz} (in units of μ_0^2/E_H where μ_0 is the Bohr magneton) as a function of T and E_0 at $n_x(a_0^*)^2 = 0.29$.

$\Delta C_v/T_c$ is not a universal ratio, i.e., larger in the strongly interacting low density BEC limit than in the weakly interacting high density BCS limit. This ratio is reduced as the SOC is increased (more rapidly at high densities) implying that the strongly second order transition is weakened by increasing the SOC. In the high density range, ΔC_v vanishes at a certain SOC strength whereas at low densities the second order transition remains but dramatically weakened. It is expected that the Hartree-Fock scheme studied here may be insufficient in explaining the details of the transition due to the strong dipolar fluctuations at high densities and near T_c. Two factors are influential on the C_v near $T \simeq 0$ and $T \simeq T_c$ when the SOC strength is increased. The first is the anisotropic narrowing of the two lowest energy exciton bands at the Fermi energy allowing low energy thermal excitations between these bands. Thermal interband excitations are effective even at $T \approx 0$ resulting in a power law dependence. The second is that the anisotropy in $\mathbf{\Delta}(\vec{k})$ persists at finite temperatures which is more affective in larger densities averaging out the anomaly in C_v more rapidly than in low densities.

Concerning $\chi_{xz}(T)$, it vanishes at all temperatures in the absence of SOC. However, it is finite in the presence of SOC [10] and stronger in low temperatures as depicted in Fig. 4. For increasing SOC, $\chi_{xz}(0)$ gradually increases and an exponential-to-power-law behavior near $T \simeq 0$ develops similar to C_v in Fig. 3(c).

In conclusion, we demonstrated three measurable effects displayed by an *e-h* system when the R&D SOCs are considered. The first is the enhancement of the BC in the ground state with observable consequences in the PL measurements. The second and the third are the nonconventional behavior of the specific heat and the spin susceptibility at $T \simeq 0$ and $T = T_c$. These three effects combined should help in the enhancement of our understanding the exciton condensation.

Recent experiments [17] have shown that the broken inversion symmetry can yield, in addition to the R&D SOCs, strain-induced spin splitting in the same order of magnitude as the SOC studied here [18]. We expect the measurements under strain to be crucially important for complementing the PL measurements.

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