Toroidal excitons in crystals

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Quasiparticles of a new type—toroidal excitons, characterized by a nonzero toroidal moment—may exist in solids. Their wave function is calculated in the effective-mass approximation.

1. Exciton states can be classified on the basis of how they behave under transformations of the group of spatial and temporal inversions. It is believed[1] that the wave function of an exciton in a nonmagnetic, centrally symmetric crystal should be either invariant under these transformations or should change sign under space inversion or time reversal. In the last two cases, the exciton can have an electric or magnetic, respectively, dipole moment. On the other hand, we know[2] that in addition to the electric and magnetic families, there is a third, and independent, family of electromagnetic multipoles: the toroidal family. We will show in this letter that a bound state of an electron and a hole having a toroidal moment—a toroidal exciton—corresponds to this family. The wave function of this state changes sign under both space inversion and time reversal.

We write the Hamiltonian of the system of electrons and holes in the form

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$$\mathcal{H} = \sum_{\mathbf{p}} (\epsilon_1(\mathbf{p}) a_{\mathbf{p}}^+ a_{\mathbf{p}} + \epsilon_2(\mathbf{p}) b_{\mathbf{p}}^+ b_{\mathbf{p}})$$

$$- \sum_{\mathbf{p},\mathbf{p}',\mathbf{Q}} \{ \frac{1}{2} [V_1(\mathbf{p}-\mathbf{p}') - 2V_2'(\mathbf{Q})] a^+_{\mathbf{p}'+\mathbf{Q}/2} b^+_{-\mathbf{p}'+\mathbf{Q}/2} b_{-\mathbf{p}+\mathbf{Q}/2} a_{\mathbf{p}+\mathbf{Q}/2}$$

$$+ [V_2(\mathbf{p}-\mathbf{p}') - 2V_2(\mathbf{Q})] a^+_{\mathbf{p}'+\mathbf{Q}/2} b^+_{-\mathbf{p}'+\mathbf{Q}/2} b_{-\mathbf{p}+\mathbf{Q}/2} a_{\mathbf{p}+\mathbf{Q}/2} + \text{H.a.} \}, \quad (1)$$

where $a_{\mathbf{p}}$, $b_{\mathbf{p}}$ are Fermi operators which annihilate an electron in the conduction band and a hole in the valence band. The dispersion law for the quasiparticles is $\epsilon_1(\mathbf{p}) = \epsilon_2(\mathbf{p}) = \mathbf{p}^2/2m^* + E_g/2$ where m^* is the effective mass, and E_g is the band gap of the crystal. In addition to the direct Coulomb interaction $V_1(\mathbf{p}) = 4\pi e^2/\kappa \mathbf{p}^2$ (e is the charge of an electron, and κ is the dielectric constant), Hamiltonian (1) incorporates the exchange coupling $V_2'(\mathbf{p})$ and the interband transitions of electrons and holes, $V_2(\mathbf{p})$. Everywhere below we set $\hbar = 1$.

The spectrum of excitons is determined by the poles of the two-particle Green's function $K(p,p',Q)$, where $Q = \{\mathbf{Q},\Omega\}$ are the resultant quasimomentum and resultant energy, and $p = \{\mathbf{p},\omega\}$ and $p' = \{\mathbf{p}',\omega'\}$ are the relative quasimomenta and energies of the electron and the hole. Since the Hamiltonian contains a term which stems from interband transitions, we need to consider, in addition to $K(p,p',Q)$, the function $\tilde{K}(p,p',Q)$, which describes the creation of two electron-hole pairs with a zero resultant quasimomentum.[3] A system of equations for these functions can be represented graphically by the diagrams shown in Fig. 1. A circle in a diagram means the sum of the irreducible diagrams of the given type. Here we have $\Sigma(p,Q) = V_1(\mathbf{p}) - 2V_2'(\mathbf{Q})$, and $\tilde{\Sigma}(p,Q)$ incorporates interband transitions.

The system of equations is solved with the help of the Green's function of the Coulomb problem.[4] Near the lowest pole we have

$$\Omega(\mathbf{Q}) = \mathbf{Q}^2/2M + E_g - E_1 + 2V_2'(\mathbf{Q}) \pm V_2(\mathbf{Q}), \quad (2)$$

where $M = 2m^*$, $E_1 = me^4/2\kappa^2$ is the binding energy of an exciton if only the $V_1(\mathbf{p})$ coupling is taken into account, and $m = m^*/2$ is the reduced mass of the electron and the pole. In this case we have the relation

$$K(p,p',Q) = \pm \tilde{K}(p,p',Q). \quad (3)$$

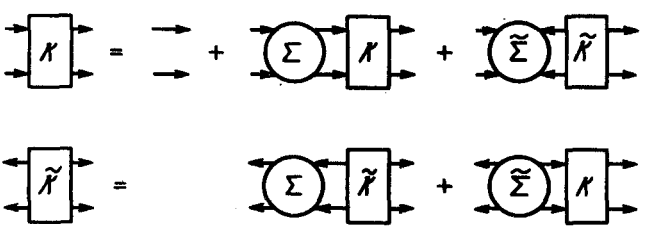

FIG. 1.

The fact that the two-particle Green's function has two types of poles indicates that there are two types of singlet bound states of the electron and hole (we are ignoring the spin degree of freedom).

2. A solution of (2) has been derived ignoring the dependence of $\tilde{\Sigma}(p,Q)$ on $K(p,p',Q)$; this simplification corresponds to the mean-field approximation. We calculate the exciton wave functions by taking this dependence into account. We write the wave function in the form

$$\Phi(\mathbf{p},\mathbf{Q}) = \phi(\mathbf{p},\mathbf{Q})\, a^{+}_{\mathbf{p}+\mathbf{Q}/2} b^{+}_{-\mathbf{p}+\mathbf{Q}/2}\, \Phi_0 , \tag{4}$$

where Φ_0 is the wave function of the ground state of the crystal. Applying Hamiltonian (1) to (4), and taking an expectation value over the internal motion, we find, for the envelope

$$\phi(\mathbf{Q}) = \int \phi_1(\mathbf{p})\phi(\mathbf{p},\mathbf{Q})\frac{d^3\mathbf{p}}{(2\pi)^3} \tag{5}$$

[$\phi_1(\mathbf{p})$ is the eigenfunction of the lowest state for the Coulomb problem], the equation

$$[\Omega - \mathbf{Q}^2/2M - E_g + E_1 - 2V_2'(\mathbf{Q})]\phi(\mathbf{Q}) \pm V_2(\mathbf{Q})|\phi(\mathbf{Q})|^2\phi(\mathbf{Q}) = 0, \tag{6}$$

where the plus sign corresponds to the case in which the function $\phi(\mathbf{Q})$ is real, and the minus sign corresponds to the case in which it is purely imaginary.

Equation (6) is of the form of a nonlinear Schrödinger equation describing a system with a variable number of particles. In the one-dimensional case, this equation has soliton solutions, which oscillate in space and time. In the coordinate representation we have[5]

$$|\phi(x,t)| = \frac{1}{2}(Mg)^{1/2}\exp[i(Qx - \Omega t)]/\cosh\left[\frac{1}{2}Mg(x - Qt/M)\right], \tag{7}$$

$$\Omega = Q^2/2M + E_g - E_1 - (Mg^2/8)$$

for an attractive point interaction $V_2(Q)\equiv g$.

3. In summary, incorporating the interband transitions of electrons and holes leads, by virtue of the $V_2(\mathbf{Q})$ coupling, to a lifting of the phase degeneracy of the wave function, i.e., the degeneracy of the exciton states under time reversal. Since in this case we have $V_2(\mathbf{Q})\neq 0$ only for the states of the conduction band and valence band which differ in parity, the degeneracy under space inversion is lifted simultaneously.

Evaluating the diagonal matrix elements of the toroidal-moment operator

$$\hat{\mathbf{T}} = \frac{-i}{20\,m^{*}c}[(\mathbf{r}(\mathbf{r}\cdot\nabla)) - 2\mathbf{r}^2\vec{\nabla}], \tag{8}$$

where c is the velocity of light, with functions (4), we verify that the density of the toroidal moment is nonzero for states characterized by a purely imaginary and spatially antisymmetric wave function.

The selection rules for transitions to toroidal exciton states are quite different from those for transitions to exciton states of a different symmetry, so there is a good opportunity for an experimental observation and identification of toroidal excitons in crystals.

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