Radiative recombination of localized Mott excitons in a strong magnetic field

V. A. Kharchenko

Leningrad Polytechnic Institute

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The effect is considered of a strong magnetic field on the formation and recombination of Mott excitons localized at defects and impurities in semiconducting crystals. It is shown that the motion of the exciton mass center across the magnetic field plays a decisive role in the formation of localized excitons. It is found that by varying the magnetic-field intensity one can control the relative brightness (radiative recombination probability) of the spectral lines of optical recombination and or absorption of bound excitons. This possibility is due to the dependence of the effective mass of the transverse motion of the exciton mass center on the magnetic field intensity.

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A Mott exciton can move with constant velocity in a direction transverse to the magnetic field even though its constituent quasiparticles, the electron and hole, cannot do so individually. As shown by Gore'kov and Dzyaloshinskii, this transverse motion of the exciton mass center is due only to the Coulomb attraction between the electron and the hole. The role of the exciton mass-center momentum is played here by the conserved quantity

\[ \mathbf{p} = -i \hbar \nabla \Psi, \]

where \( \mathbf{R} \) and \( \mathbf{r} \) are the coordinates of the mass center and of the relative motion of the quasiparticles in the exciton, and \( \mathbf{A} \) is the vector potential of the constant magnetic field. The wave functions calculated in Ref. 1 and the energies of the internal (relative) motion of the exciton depend on the projection \( \mathbf{P} \) of the vector \( \mathbf{P} \) on a direction perpendicular to the magnetic field. It is just this transverse motion, as shown in the present paper, which plays the decisive role in the formation of the bound state of the exciton with impurities and defects in strong magnetic fields. Indeed, the localized states are detached, under the influence of the defect field, from the exciton band, and their characteristics depend significantly on the "dispersion law" obtained in Ref. 1 for the exciton band in a magnetic field. The diamagnetic-exciton localization considered in this article should appear, for example, in the course of their radiative recombination, which was investigated in the absence of magnetic fields by Rashba. In our case, by changing the dispersion law of the diamagnetic excitons, one can control the relative brightness of the optical-recombination lines of the bound excitons.

The action of a defect on an electron-hole pair is taken into account within the framework of the effective-mass method by including in the Schrödinger equation the potential field \( V \) produced by the defect:

\[ \mathbf{H}_d = \frac{1}{2m_e} \left( - \nabla \Psi + \frac{\mathbf{e}}{\epsilon} \mathbf{A}_e \right)^2 + \frac{1}{2m_h} \left( - \nabla \Psi - \frac{\mathbf{e}}{\epsilon} \mathbf{A}_h \right)^2 - \frac{e^2}{\epsilon R}, \]

where \( \mathbf{H}_d \) is the Hamiltonian of the electron-hole pair in a magnetic field, and \( \mathbf{A}_e \) and \( \mathbf{A}_h \) are the vector potentials of the electron and hole, respectively. We are interested in the state of an exciton that is weakly bound with a defect, such that the defect field, while localizing the motion of the exciton mass center, does not perturb in its internal motion. In Ref. 1 were obtained for the case of strong magnetic fields \( r_0 \ll \epsilon_H (r_0 = (\hbar/eH)^{1/2} ) \) is the magnetic length and \( r_g = \hbar^2/\mu e^2 \) is the Bohr radius of the exciton) the exciton wave functions \( \Psi_{\mathbf{P}} (\mathbf{r}, \mathbf{R}) \) and exciton energy \( E_{\mathbf{P}} \):

\[ \Psi_{\mathbf{P}} (\mathbf{r}, \mathbf{R}) = E_{\mathbf{P}} (\mathbf{r}) \Psi_{\mathbf{P}} (\mathbf{r}), \]

\[ \Psi_{\mathbf{P}} (\mathbf{r}, \mathbf{R}) = \exp \left[ \frac{1}{\hbar} \int \frac{e^2}{\epsilon R} \frac{d^3 \mathbf{r'}}{R} \mathbf{A} (\mathbf{r}) \right] \Psi_{\mathbf{P}} (\mathbf{r}). \]

We seek the solution of (1) in the form of an expansion of the wave function \( \Psi_{\mathbf{P}} (\mathbf{r}, \mathbf{R}) \) in terms of the eigenfunctions of the unperturbed Hamiltonian \( \mathbf{H}_d \), (4):

\[ \Psi_{\mathbf{P}} (\mathbf{r}, \mathbf{R}) = \sum_{\mathbf{P}'} \Psi_{\mathbf{P}'} (\mathbf{r}) | \mathbf{P}' \rangle \langle \mathbf{P} | \Psi_{\mathbf{P}'} (\mathbf{r}), \]

Substituting (5) into (1) and taking the normalization of the wave functions (4) into account we obtain a system of equations for \( \Psi_{\mathbf{P}'} (\mathbf{r}) \):

\[ [E_{\mathbf{P}} (- E_{\mathbf{P}}) \delta_{\mathbf{P}^2} + \sum_{\mathbf{P}'} \Psi_{\mathbf{P}'} (\mathbf{r}) \int d^3 \mathbf{R} \exp \left[ \frac{e^2}{\epsilon R} \mathbf{A} (\mathbf{r}) \right] \frac{1}{\hbar} (\mathbf{P}' - \mathbf{P}) \mathbf{R} \rangle \times \langle \mathbf{P}' \mathbf{P} \rangle \Psi_{\mathbf{P}'} (\mathbf{r}) = 0. \]

For the cases considered by us, of excitons weakly bound to defects, the characteristic dimensions \( R \) and \( R_g \) of the mass-center motion orbit (in directions parallel and perpendicular to the magnetic field) exceed substantially the dimensions of the exciton itself: \( R \gg r_0 \) and \( R_g \gg r_0 \). Explicit expressions for \( R \) and \( R_g \) will be obtained below. It is clear that the main contribution to the formation of such an extended mass-center wave function is made by the values of \( \mathbf{P} \), namely \( P_x \ll \hbar/R \) and \( P_y \ll \hbar/R \), which are small in terms of the parameters:

\[ r_0 P_x/\hbar < 1, \quad r_0 P_y/\hbar < 1. \]
The conditions (7) mean that the states participation in the formation of the local level are located near the "bottom" of the exciton band, and leaving out of (6) the terms that are small relative to (7) and (8), we obtain an equation for the amplitude \( a_i(P) \)

\[
\{ E_i(P) - E_i(0) \} \sum_j a_j(P') \int d^2R \exp \left[ \frac{\hbar}{\hbar} (P' - P) R \right] \psi_i(R) = 0,
\]

(9)

where \( E_i(P) = E_i + \frac{\hbar^2}{2M} \), \( W_a(H) = - \frac{\hbar^2}{2M} \), and \( M = m_e + m_h \), is the effective mass of the exciton center motion in a direction transverse to the field; this will be called hereafter the magnetic mass.

Choosing for the sake of argument the case of an exciton in the ground state of the zeroth Landau band, and leaving out of (6) the terms that are small relative to (7) and (8), we obtain an equation for the amplitude \( a_i(P) \)

\[
\{ E_i(P) - E_i(0) \} \sum_j a_j(P') \int d^2R \exp \left[ \frac{\hbar}{\hbar} (P' - P) R \right] \psi_i(R) = 0,
\]

(9)

\[
\Gamma^{(i)}(r) = \int \psi_i^*(r) \psi_i(r) d^2r,
\]

(10)

is the dispersion law obtained in Ref. 1 for the ground state of an exciton in the small-momentum region \( r \), \( \hbar = 2M \) is the width of the forbidden band; \( M = m_e + m_h \), is the effective mass of the exciton center motion in a direction transverse to the magnetic field; this will be called hereafter the magnetic mass.

We rewrite (9) in a coordinate form, which is more convenient for the solution, by introducing the wave function of the exciton mass-center motion

\[
\chi(r) = \sum_i \psi_i(r) \exp \left[ \frac{\hbar}{\hbar} (P - P_i) \right],
\]

(12)

where \( E_i = E_i(0) + \frac{\hbar^2}{2M} \), \( W_a(H) = - \frac{\hbar^2}{2M} \), and \( M = m_e + m_h \), is the effective mass of the exciton center motion in a direction transverse to the magnetic field.

The total wave function of the exciton is obtained, in accordance with (5), by multiplying (13) by the internal-motion wave function

\[
\psi_i(r) \exp \left[ \frac{\hbar}{\hbar} \left( \frac{\hbar}{\hbar} r - P_i \right) \right],
\]

(13)

We find from (15) and (17)

\[
\frac{\partial}{\partial \omega} \left( \frac{\hbar^2}{2M} \right) \omega_{\text{osc}} \propto \frac{\hbar^2}{2M} \propto \frac{M^2}{2M},
\]

(18)

where \( \omega_{\text{osc}} \) is the frequency of the oscillator.

The probability \( \Gamma^{(i)} \) of the radiative recombination of a bound exciton, per unit defect is

\[
\Gamma^{(i)}(r) = \int \left[ \psi_i^*(r) \psi_i(r) \right] d^2r,
\]

(14)

Thus, the dependence of the external motion of an exciton in a magnetic field on the translational motion of its mass center has appeared in (12) as the dependence of the transverse-motion mass on the magnetic field intensity.

For a weakly bound exciton the solution of Eq. (12) outside the range of action of the defect potential is of the form

\[
\chi(r) = \exp \left[ - \frac{r^2}{2G} \right],
\]

(15)

where

\[
\Gamma^{(i)} = \left( \frac{\hbar^2}{2M} \right) \omega_{\text{osc}} \propto \frac{\hbar^2}{2M} \propto \frac{M^2}{2M},
\]

(16)

It can be seen from (13) that the characteristic dimensions of the exciton mass-center orbit are determined by binding energy \( E_b(H) \) of the mass center with the defect:

\[
R_s = \left( \frac{M}{2M_s} \right)^{1/2}, \quad R_e = \left( \frac{M}{2M_s} \right)^{1/2},
\]

(17)

where \( R_s \) is the magnetic field intensity at which \( r_s = r_s \). The probabilities \( \Gamma^{(i)} \) and \( \Gamma^{(ii)} \) are proportional to the oscillator strengths \( f_{ij} \) and \( f_{ij} \) of the considered optical transitions, and the relative brightness of the luminescence lines of the bound and free excitons depends on the magnetic field like

\[
\frac{\Gamma^{(i)}}{\Gamma^{(ii)}} \propto \frac{f_{ij}}{f_{ij} f_{ij}} \propto \frac{M^2}{2M} \propto \frac{M^2}{2M},
\]

(18)

This behavior of the relative brightness has a simple physical explanation. Following Rashba, we find from (15) and (17) that the relative brightness

\[
\frac{\Gamma^{(i)}}{\Gamma^{(ii)}} = \left( \frac{\hbar^2}{2M} \right) \omega_{\text{osc}} \propto \frac{M^2}{2M},
\]

(19)

where

\[
\omega_{\text{osc}} = \left( \frac{\hbar^2}{2M} \right) \omega_{\text{osc}} \propto \frac{M^2}{2M},
\]

(16)

It should be noted that \( M \) is the magnetic field intensity at which the characteristic dimensions of the exciton mass-center orbit are determined by binding energy \( E_b(H) \) of the mass center with the defect:

\[
R_s = \left( \frac{M}{2M_s} \right)^{1/2}, \quad R_e = \left( \frac{M}{2M_s} \right)^{1/2},
\]

(17)

where \( R_s \) is the magnetic field intensity at which \( r_s = r_s \). The probabilities \( \Gamma^{(i)} \) and \( \Gamma^{(ii)} \) are proportional to the oscillator strengths \( f_{ij} \) and \( f_{ij} \) of the considered optical transitions, and the relative brightness of the luminescence lines of the bound and free excitons depends on the magnetic field like

\[
\frac{\Gamma^{(i)}}{\Gamma^{(ii)}} \propto \frac{f_{ij}}{f_{ij} f_{ij}} \propto \frac{M^2}{2M} \propto \frac{M^2}{2M},
\]

(18)

This behavior of the relative brightness has a simple physical explanation. Following Rashba, we find from (15) and (17) that the relative brightness

\[
\frac{\Gamma^{(i)}}{\Gamma^{(ii)}} = \left( \frac{\hbar^2}{2M} \right) \omega_{\text{osc}} \propto \frac{M^2}{2M},
\]

(19)
is determined by the characteristic volume of the region of motion of the mass center of the localized exciton; this volume is

\[ R_x^3 R_y^3 = \text{const} \cdot M_0 R_x. \]

The increase of the magnetic mass with increasing magnetic field strength \( M_x = H \ln^{-3} \) (see \( M_x \)) causes a corresponding decrease of the volume \( R_x^3 R_y^3 \). Thus, according to (19), the relative brightness of the recombination line of the bound excitons decreases with increasing \( H \), against the background of the increase of the recombination probability of the free-exciton luminescence line (17). This increase of the probability \( P_n^F \sim H \ln (H / H_c) \) of free-exciton recombination offsets almost completely the decrease of the volume \( R_x^3 R_y^3 \), due to the increase of the magnetic mass, and the absolute brightness of the bound-exciton luminescence lines does not depend explicitly on \( H \):

\[ P_b = P_b(0) + \frac{R_x^3 R_y^3}{P^F} \sim \text{const} \cdot H \ln \left( \frac{H}{H_c} \right). \]

We note that such a compensation is possible only for the analytic form obtained in Ref. 1 for the \( M_x(H) \) dependence.

The increase of the transverse (magnetic) mass with increasing magnetic-field intensity creates more favorable conditions for the localization of the exciton mass center on defects. It follows therefore that the defects, which are incapable of capturing an exciton in the absence of a magnetic field, can localize the exciton in a sufficiently strong magnetic field \( H > H_c \), where \( H_c \) is the critical value of the field needed for the onset of a bound state. Let us estimate its value. The conditions for the onset of a bound state in a perturbing potential field were discussed in the book by Landau and Lifshitz for the case of isotropic masses. The Schrödinger equation (12) for the wave function of the mass center can be reduced to the standard isotropic form by deforming the coordinate axes. The dimensionless parameter that determines the possibility of the appearance of discrete level in the field of a defect,

\[ T = T(H) = \frac{M_x(H)}{2 \pi R^2} \left\{ \int_0^\infty \frac{\rho^2}{P^F} d\rho \right\}, \]

depends on the magnetic field intensity. If \( T \geq 1 \), the condition for the appearance of discrete level can be fulfilled. From the last condition we obtain an estimate of the critical field \( H_c \).

\[ H_c = \left( M_x / M_0 \right) T^{-1} \rho_0, \]

where \( T \) is the corresponding localization parameter at \( H = 0 \). Thus, at \( H > H_c \), there appear localized states of the defects that cannot hold on to an exciton in weaker magnetic fields.

The appearance of a bound-exciton luminescence line in the immediate region of appearance of a level at \( H > H_c \), can be perceived as an additional broadening of the radiative-recombination line of the free excitons. In fact, a small increase of the magnetic mass with increasing \( H \) is equivalent to an increase of the depth of the potential well of the defect (see (20)). Then, in analogy with Ref. 4, we can estimate the behavior of \( I(H) \) near the level-appearance threshold at \( H = H_c \).

\[ I(H) = \text{const} (H - H_c)^3. \]

In this case, according to (15), the brightness (recombination probability) of the considered bound-exciton recombination line decrease with increasing field like

\[ P_b = \text{const} (H - H_c)^2. \]

Further increase of \( H \) should lead to a “detachment,” from the free-exciton recombination line, of localized-exciton luminescence spectral lines whose oscillator strengths (15) are large because of the sufficiently low detachment energies \( I(H) \).

We establish now the character of the dependence of \( I \) on the magnetic-field intensity \( H \) at \( H > H_c \). The large difference between the masses of the longitudinal and transverse mass-center motion, \( M_x / M_y < \ll 1 \), permits a “adiabatic” separation of these motions. The longitudinal-motion wave function \( \chi_z (z, \rho) \) (p plays the role of a parameter) satisfies a one-dimensional equation obtained from (12) in the zeroth approximation in \( M_x / M_y \rightarrow \infty \):

\[ \left\{ \begin{array}{l}
- \frac{H^2}{2M_y} \frac{\partial^2}{\partial z^2} + V(z, \rho) \chi_z(z, \rho) = \epsilon_z(p) \chi_z(z, \rho).
\end{array} \right. \]

(22)

\( \epsilon_z(p) \) will be the potential energy for the two-dimensional transverse-motion equation obtained when account is taken of the next terms in \( M_y / M_x \) of Eq. (12) averaged over the zeroth-approximation wave function \( \chi_z (z, \rho) \):

\[ \left\{ \begin{array}{l}
- \frac{H^2}{2M_y} \frac{\partial^2}{\partial z^2} + V(z, \rho) \chi_z(z, \rho) = \epsilon_z(p) \chi_z(z, \rho).
\end{array} \right. \]

(23)

For a weak potential \( V(z, \rho) \) that does not bind the exciton mass center without a magnetic field, the energy of the one-dimensional longitudinal motion is according to Ref. 3

\[ \epsilon_z(p) = - \frac{M_y}{2R} \left( \int_0^R V(z, \rho) dz \right). \]

(24)

There is no transverse motion as \( M_y \rightarrow \infty \). For large but finite values of \( M_y / M_x \), the exciton mass center executes small transverse oscillations about the equilibrium position, to which the lowest value of \( \epsilon_z(p) \) corresponds. We consider for simplicity a spherically symmetrical potential \( V \) with a minimum at \( \rho = 0 \). Expanding \( \epsilon_z(p) \) in (23) in powers of \( \rho \) and retaining the quadratic term, we obtain an equation for a planar harmonic oscillator displaced in energy by an amount \( e_z = \epsilon_z(0) \) (24) and having an oscillation frequency \( \omega_z(H) \):

\[ \omega_z^2(H) = \frac{1}{M_y} \left\{ \int_0^R V(z, 0) \left[ \frac{\partial^2}{\partial \rho^2} V(z, \rho) \right] \right\}. \]

(25)

Then

\[ I(H) = |e_z| - \hbar \omega_z(H). \]

(26)

It follows from (25) that

\[ \hbar \omega_z = |e_z| (H / M_x R)^2 \ll |e_z| \]

at \( H > H_c \). In the limit of an infinitely large magnetic mass...
\[ M_e(H \to -\omega) \] we have \( \kappa_{\omega} \to 0 \) and the binding energy \( J(\omega) \) tends to the binding energy \( E_0 \) of the ground state of the one-dimensional motion. Thus, for arbitrary \( H \),

\[ I(\omega) \ll |E_0| \ll E_B. \]

At \( H \approx H_c \), the wave function \( \chi_{\omega}(p) \) is strongly localized because of the large magnetic mass \( M_0 \). The characteristic scale \( R_{\omega} \) of the variation of the transverse wave function is not determined as in (14a) by the binding energy \( I - |E_0| \), but depends on the energy \( \kappa_{\omega} \) of the transverse oscillations:

\[ R_{\omega} = \left( \frac{M}{\kappa_{\omega}} \right)^{1/2} \left( \frac{2E_0 \kappa_{\omega}}{M_0} \right)^{1/2} \]

(27)

with \( \kappa_{\omega} \to 0 \) as \( H \to -\omega \). Despite the strong localization we have \( R_{\omega} \gg \ell_{\omega} \), and the condition for the applicability of the parabolic dispersion law (7)

\[ \left( \frac{\ell_{\omega} R_{\omega}}{\ell_{\omega}} \right) = \frac{\hbar \omega}{2E_0 \kappa_{\omega}} < 1 \]

remains valid also in the case of very strong fields \( H \approx H_c \).

Using the explicit forms of \( \chi_{\omega} \) and \( \chi_{\omega} \), we obtain the radiative-recombination probability \( \mathcal{P}_{\omega} \) as \( H \to -\omega \):

\[ \mathcal{P}_{\omega} = \frac{\lambda_{\omega}}{\hbar^2 g_{\omega}^2 \ell_{\omega}^2} \left( \frac{2E_0 \kappa_{\omega}}{M_0} \right)^{1/2} \]

(28)

where \( \lambda_{\omega} \sim |E_0|/|J(H_c - \omega)| = |J|^{1/2} \). The square-root dependence on \( H \) in (28) is due to the dependence of \( R_{\omega} \) on the transverse-oscillation energy \( \kappa_{\omega} \):

\[ \mathcal{P}_{\omega} \propto \left( \frac{R_{\omega}}{R_{\omega}} \right)^{1/2} \propto \omega^{-1/2} \]

the longitudinal dimension \( R_{\omega} \) and the binding energy \( I - |E_0| \) are practically independent of \( H \) as \( H \to -\omega \).

The decrease of \( M_0 \) (or of \( H \)) leads to an increase of the energy \( \kappa_{\omega} \) of the transverse oscillations. Clearly, at \( H \approx H_c \), \( \kappa_{\omega} \sim |E_0| \) the transverse motion becomes "weakly bound" and, according to Ref. 3, the energy of such a state is exponentially small. The adiabatic approximation (22), (23) is therefore violated in the immediate vicinity \( H \approx H_c \). The longitudinal and transverse motions exert a strong influence on each other. The foregoing estimates show that the scale of variation of \( I \) at \( H_c < H < \infty \) is determined by the binding energy of the one-dimensional motion of the exciton mass center, and the functional form of \( I(\omega) \) is given by Eqs. (21) and (26).

The considered characteristic features of the radiative recombination of diamagnetic excitons in the presence of defects can be observed only at low temperatures, \( \leq 10 \) K. The critical values \( H_c \) of the magnetic fields, in a situation typical of the observation of diamagnetic excitons, can reach values from \( 10^2 \) to \( 10^3 \) kOe, depending on the characteristic of the perturbing field of the defect.

All the foregoing results and estimates are applicable also to the case of localization of excited diamagnetic excitons. Owing to the large magnetic mass of such excitons, \( M_0 \propto v^2 M_0 \) (where \( v \) is the number of the excited state) their localization should set in at lower values of the critical fields: \( H_c \approx v^2 H_c \).

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1. Accurate to a logarithmic term \( \Delta \sim -\ln(\omega/\omega_0) \).

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