

Negatively Charged Exciton on a Quantum Ring

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Dedicated to Professor Dr. Roland Zimmermann on the occasion of his 60th birthday

We present calculations of the magnetic field dependence of the emission spectrum of charged exciton (X^-) localised on a quantum ring. They are carried out using exact diagonalisation techniques in a coordinate system localised on a valence hole. The ground state energy of charged X^- complex, unlike neutral exciton, exhibits Aharonov–Bohm oscillations for any disk radius. The emission energy is found as a difference of Aharonov–Bohm oscillations in the charged exciton and the final state electron energy.

Introduction The evolution of the optical properties of low dimensional semiconductor structures with carrier density has been extensively studied [1–13]. It is now well recognised that the low-density regime is dominated by the exciton and charged exciton absorption/recombination. The understanding of the role of negatively charged exciton, including dynamical processes, has been advanced largely by the work of Zimmermann and co-workers [12]. Our own discussion of negatively charged exciton was motivated by the attempt to understand the approach to the high density limit dominated by Fermi edge singularity [4]. In this limit the role of finite valence hole mass in the emission/absorption is a serious and unsolved problem [4]. A possible solution to the problem lies in the coordinate system where hole mass can be treated perturbatively. We studied such a coordinate system in the simplest case of a two-dimensional X^- [13]. Motivated by recent experiments on charged excitons in quantum rings [14] we extend the approach of Ref. [13] to quantum rings and to magnetic fields.

In quantum ring geometry the phase of the single electron wavefunction changes with the external magnetic field. This results in oscillations of the electronic energy as a function of the number of magnetic flux quanta threading the ring, the Aharonov–Bohm (AB) oscillations [15]. The possibility of observing Aharonov–Bohm oscillations in quantum rings was studied experimentally [16–18] and theoretically. Theoretical work started with Chaplik and Govorov [19] who studied excitons and charged exciton complexes. Finite size effects of quantum rings on magneto-excitons were also extensively studied [20–22]. It appears that in spite of its charge neutrality, the low-lying energy levels and oscillator strengths of an exciton also exhibit the AB oscillations as a function of the magnetic field, provided that the ring is small and narrow.

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Chaplik [19] considered a system of $N + 1$ electrons and a hole, or the X^{N-} complex, localized on a ring with small, but finite thickness. He finds that in this case the binding energy of the multiply-charged complex exhibits a negative diamagnetic shift (due to the radial degrees of freedom) and AB oscillations, whose period depends on the ratio of effective masses of electrons and the hole. Romer and Raikh [23] add to this description a detailed analysis of electronic spins, distinguishing an ortho- and para-trion (electronic triplet and singlet, respectively), but only at zero magnetic field.

Our analysis is similar to that of Refs. [13, 19, 23]. We assume the quantum ring to be infinitesimally thin (one-dimensional), and use modified Coulomb interactions. We use exact diagonalisation techniques to calculate exciton and charged exciton energy levels, and recombination spectra as a function of the magnetic field. Both spin triplet and spin singlet trions were studied but spin singlet configurations were found to be the lowest energy states and they are discussed here. This approach captures the essential physics of the system, and allows to analyse systematically the influence of the ring size on the period and magnitude of the AB oscillations of X^- complex. However, the introduction of a special set of relative coordinates in the solution of the problem allows additionally for a straightforward analysis of the influence of the hole effective mass on the magneto-photoluminescence spectra of the system.

Model We consider the X^- complex: two electrons, each with mass m_e and charge $-e$, and one hole with mass m_h and charge $+e$, localised on a quantum ring. An external magnetic field is applied perpendicularly to the plane of the ring. The Hamiltonian of two electrons and one hole is given by

$$\hat{H} = \frac{\left(\mathbf{p}_1 + \frac{e}{c}\mathbf{A}_1\right)^2}{2m_e} + \frac{\left(\mathbf{p}_2 + \frac{e}{c}\mathbf{A}_2\right)^2}{2m_e} + \frac{\left(\mathbf{p}_h - \frac{e}{c}\mathbf{A}_h\right)^2}{2m_h} - V_{eh}(\mathbf{r}_1, \mathbf{r}_h) - V_{eh}(\mathbf{r}_2, \mathbf{r}_h) + V_{ee}(\mathbf{r}_1, \mathbf{r}_2), \quad (1)$$

where \mathbf{r}_i are the electron coordinates, \mathbf{r}_h is the valence hole coordinate, and $\mathbf{A}_i, \mathbf{A}_h$ are the magnetic vector potentials for each electron and the hole, respectively. The potential $V_{eh}(\mathbf{r}_e, \mathbf{r}_h)$ describes the attractive electron–hole Coulomb interaction, and $V_{ee}(\mathbf{r}_1, \mathbf{r}_2)$ the repulsive electron–electron interaction. In what follows we assume that the ring has radius R and infinitesimal thickness, so that the electronic coordinates can be written as $\mathbf{r}_i = (R \cos \theta_i, R \sin \theta_i)$, and the hole coordinates are $\mathbf{r}_h = (R \cos \theta_h, R \sin \theta_h)$. We shall analyse the Hamiltonian (1) systematically term by term.

We start with the kinetic energy operators. Since the momentum operator for each particle involves only differentiation over the respective angle, the kinetic energy operator for each electron can be written as

$$\hat{T}_e = \frac{\hbar^2}{2m_e R^2} \left(-i \frac{\partial}{\partial \theta} + N_\phi \right)^2, \quad (2)$$

where $N_\phi = \pi R^2 / 2\pi l^2$ is the number of flux quanta threading the ring, $l = \sqrt{\hbar / m_e \omega_c}$ is the magnetic length, and $\omega_c = eB / m_e c$ is the cyclotron frequency (we take the magnetic potentials in the symmetric gauge). The kinetic energy operator for the hole takes an analogous form, only m_e is replaced by m_h , and the number of flux quanta N_ϕ enters with negative sign due to the positive charge of the hole. The eigenstates of this single-

particle kinetic energy operator are $\psi_k(\theta) = 1/\sqrt{2\pi} e^{ik\theta}$, where k is the angular momentum quantum number, and the corresponding eigenenergies are $E_k^e = \frac{\hbar^2}{2m_e R^2} (k_e + N_\phi)^2$ for each electron, and $E_k^h = \frac{\hbar^2}{2m_h R^2} (k_h - N_\phi)^2$ for the hole. Thus, the kinetic energy of a particle with a definite angular momentum depends quadratically on the number of flux quanta. The low energy sector as a function of N_ϕ will then consist of a set of cut parabolas, with smooth minima for integer N_ϕ , and sharp maxima for half-integer N_ϕ , for which the angular momentum of the single-particle ground state changes by one unit (becomes more negative for electrons, and more positive for the hole). The ground state energies of the single electron and the single hole localised on the ring are shown in Fig. 1. The unit of energy is $\hbar^2/2m_e R^2$, and the ratio $m_e/m_h = 0.25$. The amplitude of oscillations of the single-particle ground state energy is smaller for the hole due to its larger mass.

In order to include interactions we adopt the following approximation for the Coulomb electron–hole potential

$$V_{eh}(\mathbf{r}_e, \mathbf{r}_h) = \frac{e^2}{2\varepsilon} \frac{1}{\sqrt{D^2 + R^2 \sin^2\left(\frac{\theta_e - \theta_h}{2}\right)}}, \quad (3)$$

where ε is the dielectric constant of the material, and the parameter D was introduced to account for the finite thickness of the ring and to prevent the Coulomb interaction from diverging. The electron–electron term is approximated in an analogous way. Let us consider the influence of the electron–hole interaction first. To this end it is convenient to simplify the system to just one electron and a hole, or a single exciton localised on the ring. Therefore we exclude from the Hamiltonian (1) the second term (the kinetic energy of the second electron) and the two last interaction terms. Further we introduce the center of mass (CM) and relative coordinates

$$\phi_{CM} = \frac{m_e}{M_X} \theta_e + \frac{m_h}{M_X} \theta_h, \quad (4)$$

$$\phi_{rel} = \theta_e - \theta_h. \quad (5)$$

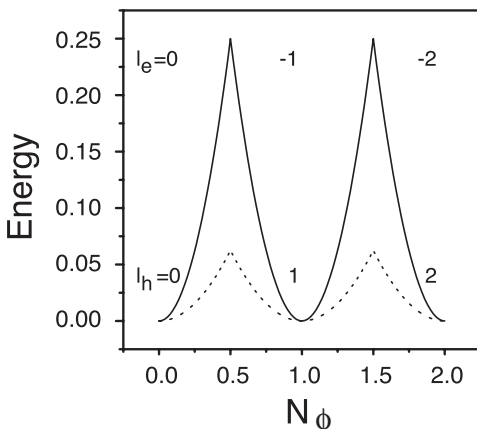


Fig. 1. Ground state energy of a single electron (solid line) and a single hole (dashed line) localised on the ring as a function of the number of flux quanta. The unit of energy is $\hbar^2/2m_e R^2$, where R is the radius of the ring

Here $M_X = m_e + m_h$ is the total mass of the exciton. The exciton Hamiltonian in these new coordinates separates into the CM and relative part

$$\hat{H}_X = \hat{H}_{\text{CM}} + \hat{H}_{\text{rel}}, \quad (6)$$

$$\hat{H}_{\text{CM}} = -\frac{\hbar^2}{2M_X R^2} \frac{\partial^2}{\partial \phi_{\text{CM}}^2}, \quad (7)$$

$$\hat{H}_{\text{rel}} = \frac{\hbar^2}{2\mu R^2} \left(-i \frac{\partial}{\partial \phi_{\text{rel}}} + N_\phi \right)^2 - \frac{e^2}{2R\epsilon} \frac{1}{\sqrt{d^2 + \sin^2(\phi_{\text{rel}}/2)}}, \quad (8)$$

where $\mu = m_e m_h / (m_e + m_h)$ is the reduced exciton mass, and $d = D/R$. As can be seen, the CM particle is charge neutral, and its energy $E_{\text{CM}}(K) = \hbar^2 K^2 / 2M_X R^2$ does not depend upon the magnetic field. The CM ground state energy corresponds to the CM angular momentum $K = 0$ and equals zero. Therefore only the relative Hamiltonian is of interest. Its form suggests that the natural units of energy and length are the excitonic Rydberg $\mathcal{R}^\mu = \mu e^4 / 2\epsilon \hbar^2$, and the excitonic Bohr radius $a^\mu = e^2 \hbar^2 / \mu \epsilon^2$, respectively, which we shall use henceforth. Note that in the Hamiltonian (8) the kinetic energy is proportional to R^{-2} , and the interaction term to R^{-1} . Thus changing the ring radius changes the ratio between the kinetic and Coulomb energies and transfers the system from weakly (small radii) to strongly interacting (large radii). We shall now carry out an exact diagonalisation study of the relative Hamiltonian in the basis of single-particle states

$$\langle \mathbf{r} | k \rangle = \frac{1}{\sqrt{2\pi}} e^{ik\phi_{\text{rel}}}. \quad (9)$$

In the basis of these states we construct the relative exciton Hamiltonian matrix and diagonalise it numerically. To this end we need the matrix element

$$\langle k_2 | V_{\text{eh}} | k_1 \rangle = \frac{1}{R} \int_{-\pi}^{\pi} \frac{dx}{2\pi} \frac{\cos(k_1 - k_2)x}{\sqrt{d^2 + \sin^2(x/2)}}. \quad (10)$$

This matrix element mixes states with different angular momenta. This allows us to predict that in the regime of weak interactions (small R) the exciton will exhibit a behaviour similar to that of the single particle (cf. Fig. 1), i.e., oscillations of the ground state energy as a function of the number of flux quanta. In the limit of strong interactions, however, the Coulomb term will mix configurations, which will result in a smoothing-out of the magnetic oscillations of the exciton energy. We performed numerical calculations for the basis set with $k = -10, -9, \dots, 10$, and for three different radii $R = 0.2, 0.7$ and $1 a^\mu$ (the “ring thickness” $D = 0.05 a^\mu$ was kept constant). The result is presented in Fig. 2, where we plot the corresponding exciton energies multiplied by R^2 , which removes the dependence of the kinetic energy on the ring radius. As predicted, in the case of weak interactions the magnetic field dependence of the ground state energy exhibits distinct oscillations and resembles that of a single particle, but as we increase the radius, the configurations with different angular momenta are mixed, and in the limit of large R the exciton energy depends smoothly on the number of flux quanta, exhibiting the diamagnetic shift only.

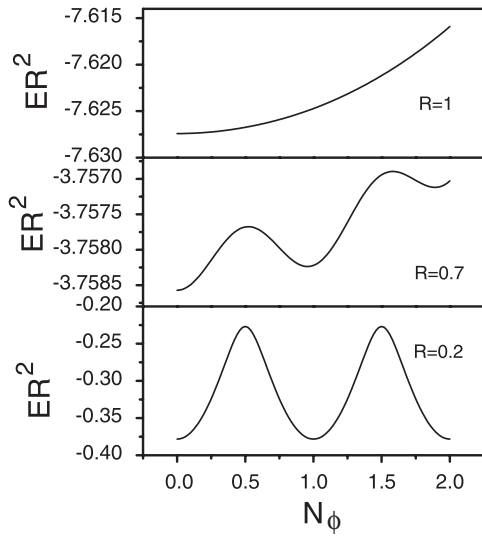


Fig. 2. Ground state energy of an exciton localised on the ring as a function of the number of flux quanta for different ring radii: $R = 1$ (strong electron-hole interactions), 0.7 and $0.2a^H$ (weak electron-hole interactions)

Let us now extend this simple analysis to account for the second electron. We introduce a set of the angular CM and relative coordinates in such a way that the coordinates of relative particles are just those of electrons but measured with respect to the hole [13]

$$\phi_{\text{CM}} = \frac{m_e}{M} \theta_1 + \frac{m_e}{M} \theta_2 + \frac{m_h}{M} \theta_h, \quad (11)$$

$$\phi_i = \theta_i - \theta_h, \quad i = 1, 2 \quad (12)$$

where $M = 2m_e + m_h$ is the total mass of the X^- complex. Upon transformation of the Hamiltonian ([1]) into the new coordinate system the CM motion separates and the new total Hamiltonian can be written as a sum

$$\hat{H} = \hat{H}_{\text{CM}} + \hat{H}_{\text{rel}}, \quad (13)$$

$$\hat{H}_{\text{CM}} = \frac{\hbar^2}{2MR^2} \left(-i \frac{\partial}{\partial \phi_{\text{CM}}} + N_\phi \right)^2, \quad (14)$$

$$\begin{aligned} \hat{H}_{\text{rel}} = & \sum_{i=1}^2 \frac{\hbar^2}{2\mu R^2} \left(-i \frac{\partial}{\partial \phi_i} + N_\phi \right)^2 - \frac{e^2}{2R\epsilon} \frac{1}{\sqrt{d^2 + \sin^2(\phi_i/2)}} \\ & + \frac{e^2}{2R\epsilon} \frac{1}{\sqrt{d^2 + \sin^2((\phi_1 - \phi_2)/2)}} + \frac{\hbar^2}{m_h R^2} \left(-i \frac{\partial}{\partial \phi_1} \right) \left(-i \frac{\partial}{\partial \phi_2} \right) \\ & - \frac{2\sigma}{1 + 2\sigma} \frac{\hbar^2}{2\mu R^2} N_\phi^2, \end{aligned} \quad (15)$$

where $\sigma = m_e/m_h$. The CM Hamiltonian describes a charged particle with the mass M localised on the ring, and the relative Hamiltonian is that of two charged particles each with mass of the exciton μ , but with the charge of an electron, moving in the attractive potential of a localised point charge. The relative particles interact via Coulomb interactions V_{ee} and a pairwise momentum interaction term V_{pp} . The pairwise interaction term is inversely proportional to the hole mass and can be treated as a perturbation when the hole is very heavy. Neglecting the momentum interaction reduces the relative Hamiltonian to the problem of two electrons in the field of a positive charge, i.e., the D^- problem on a ring. For a finite hole mass the X^- relative Hamiltonian depends on two masses, the exciton mass μ and the hole mass m_h . Let us first analyse the CM Hamilto-

nian, which describes now the charged particle. We use consistently the excitonic units of energy \mathcal{R}^μ and length a^μ . The CM eigenenergies, analogous to the single-particle energies, are

$$E_{\text{CM}}(K) = \frac{\sigma}{(2\sigma + 1)(\sigma + 1)} \frac{1}{R^2} (K + N_\phi)^2, \quad (16)$$

where K is the CM angular momentum, and the corresponding wave functions are $\psi_K(\phi_{\text{CM}}) = \sqrt{1/2\pi} e^{iK\phi_{\text{CM}}}$. The ground state energy of the CM particle behaves analogously to the single-particle energy, with smooth minima at integer, and sharp maxima at half-integer number of flux quanta, however the amplitude of these oscillations is now even smaller than that in the case of the single hole due to larger X^- mass M . This behaviour is fundamentally different than that of the CM of the exciton, whose ground state CM energy does not depend upon the magnetic field.

In the relative X^- Hamiltonian, the dependence on the magnetic field is manifested only in the kinetic energy. Moreover, similarly to the case of the single exciton, by changing the ring radius we can change the regime from strongly interacting (large R) to weakly interacting (small R). To solve for eigenenergies and eigenvectors of H_{rel} we employ also numerically exact diagonalisation. To construct the basis of the Hilbert space we first note that the total spin of the relative particles is a good quantum number. Therefore the basis two-particle wave functions for the singlet are taken as space-

symmetric functions $\psi_{k_1, k_2}^S(\phi_1, \phi_2) = \frac{1}{\sqrt{2}}(\psi_{k_1}(\phi_1) \psi_{k_2}(\phi_2) + \psi_{k_1}(\phi_2) \psi_{k_2}(\phi_1))$ if $k_1 \neq k_2$, and $\psi_{k_1, k_2}(\phi_1, \phi_2) = \psi_{k_1}(\phi_1) \psi_{k_2}(\phi_2)$ if $k_1 = k_2$, and for the triplets – the space-antisymmetric functions $\psi_{k_1, k_2}^T(\phi_1, \phi_2) = \frac{1}{\sqrt{2}}(\psi_{k_1}(\phi_1) \psi_{k_2}(\phi_2) - \psi_{k_1}(\phi_2) \psi_{k_2}(\phi_1))$ (for $k_1 \neq k_2$ only).

We can now proceed to construct the Hamiltonian matrix. The kinetic part of the Hamiltonian (15) conserves the total angular momentum of the pair of relative particles. The energies of the two-particle states oscillate with the magnetic field and are infinitely degenerate, as we can construct e.g. a state with total angular momentum 0 in infinitely many ways. Inclusion of the interaction part of the Hamiltonian requires us to define one- and two-body Coulomb matrix elements. The one-body element is analogous to that defined for the exciton (see Eq. (10)), supplemented by the suitable delta function for the second relative particle in the two-particle wave function. As for the two-body matrix elements, they fall into two categories: the two-body Coulomb matrix elements $\langle k_1, k_2 | V_{\text{ee}} | k_3, k_4 \rangle$ are

$$\langle k_1, k_2 | V_{\text{ee}} | k_3, k_4 \rangle = \frac{1}{R} \delta_{k_1+k_2, k_3+k_4} \times \int_{-\pi}^{\pi} \frac{dx}{2\pi} \frac{\cos(k_2 - k_3)x}{\sqrt{d^2 + \sin^2(x/2)}}, \quad (17)$$

and the two-body momentum matrix elements $\langle k_1, k_2 | V_{\text{pp}} | k_3, k_4 \rangle$ are

$$\langle k_1, k_2 | V_{\text{pp}} | k_3, k_4 \rangle = \frac{1}{R^2} \frac{2\sigma}{1 + \sigma} \delta_{k_1, k_4} \delta_{k_2, k_3} k_3 k_4. \quad (18)$$

Note that both two-particle matrix elements conserve the total angular momentum of the pair, $k_1 + k_2 = k_3 + k_4$. Their presence will lead to removal of the infinite degeneracy of eigenenergies in each angular momentum subspace, but it cannot prevent the

energies from oscillating, as the Coulomb interactions do not depend on the magnetic field. Therefore if we neglect the electron–hole interactions (i.e., account for only electron–electron and momentum–momentum terms) we expect sharp maxima of energies and transitions to new ground states (with total angular momenta more and more negative) for each magnetic field corresponding to a half-integer number of flux quanta threading the ring. The situation changes upon inclusion of the single-body Coulomb matrix element. This element leads to mixing of states with adjacent angular momenta, as it did in the case of the exciton. As the contribution of the Coulomb interactions to the energy of the system depends upon the ring radius, we expect to see small angular momentum mixing and large relative energy oscillations for small R , and large mixing and smooth relative energy dependence for large R as a function of the number of flux quanta.

For our numerical procedure we build the X^- relative wave function both in the singlet and the triplet subspaces in terms of $N = 231$ two-particle states generated on a set of 21 single-particle orbitals with $k = -10, -9, \dots, +10$. The calculations carried out for several ring radii indicate that within this basis the ground state is of the singlet character, so in what follows we focus on this spin subspace only. Figure 3 shows the calculated ground state energies of the relative X^- Hamiltonian multiplied by R^2 for three different ring radii $R = 1, 2.5$, and $3 a^H$ as a function of the number of flux quanta.

In order to examine its influence on the behaviour of the system, the “ring thickness” D was taken to be larger than in the case of the exciton: $D = 0.25 a^H$. As expected, we find that the overall magnetic field evolution of the relative ground state energy is similar to that of a single exciton: for small radii we find oscillations of the energy, whereas for large radii it depends smoothly on N_ϕ . Note, however, that the traces qualitatively similar to those of a single exciton are now observed for larger radii. This is due to the fact that in the present calculation the parameter D was increased by a factor of 5 (i.e., we simulate a “thicker” ring), which shifts the onset of the strong interaction limit to larger R . One important difference between the excitonic and

X^- relative energies is that for an exciton the minima, if observed, correspond to integer flux quanta, as it is the case for a single-particle spectrum. However, in the case of the X^- , the number of flux quanta corresponding to the energy minimum is not integer, and depends upon

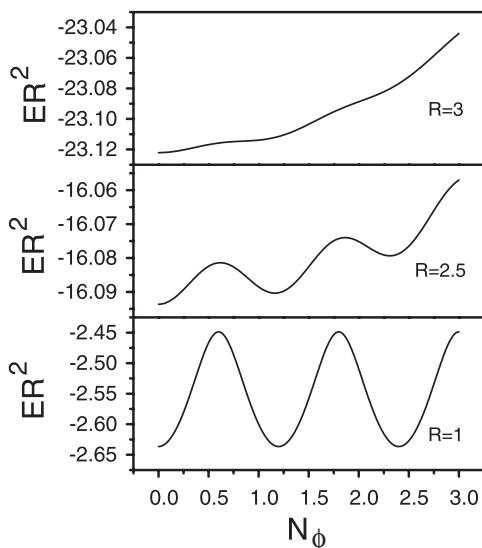


Fig. 3. Ground state energy of the relative Hamiltonian describing a negatively charged exciton localised on the ring as a function of the number of flux quanta for different ring radii: $R = 3$ (strong electron–hole interactions), 2.5 and $1a^H$ (weak electron–hole interactions)

the disk radius. This can be due only to the two-body electron–electron term, as the function of the single-body term is the same as in the case of the exciton, and the dependence of the momentum-momentum term on radius is removed by plotting the energy multiplied by R^2 .

The energy structure of both exciton and the X^- complex can be probed, e.g., by a magneto-photoluminescence experiment. The energy of the emitted photon ω is then equal to the difference between the initial and final states of the system: $\omega = E_i - E_f$. In the case of exciton the CM energy is zero, independently of the magnetic field, and the final state is vacuum. It means that the energy of the observed photon is simply equal to the ground state energy of the relative excitonic Hamiltonian, shown in Fig. 2. The magnetic oscillations in the excitonic spectrum of the ring will then be visible only if the ring radius is small enough.

For the X^- complex the energy of the initial state is the sum of energies of the CM and relative motions. The CM energy oscillates with the magnetic field for all ring radii, and the oscillations of the relative energy become smaller as R increases. Therefore the total energy of the X^- exhibits magnetic oscillations. The final state is a single electron, whose energy also oscillates with the magnetic field, however the amplitude of these oscillations is larger than it is for X^- because of the small electron mass. Therefore the photon energy $\omega = E(X^-) - E(1)$ will exhibit strong magnetic oscillations, due predominantly to the final state (electron) energy.

Figure 4 shows the result of our calculation. In this case we take $D = 0.1 a^\mu$ and $R = 2 a^\mu$. For a typical effective electron mass $m_e = 0.05 m_0$ and the ratio $\sigma = 1/4$ this radius corresponds to about 32 nm. As we can see, in this regime the relative energy of the X^- depends smoothly on the number of flux quanta, showing the diamagnetic shift only. The inclusion of the CM energy leads to the appearance of the upward cusps in total energy of the X^- . However, the photon energy is dominated by the single electron energy $E(1)$, responsible for the sharp minima at half-integer flux quanta.

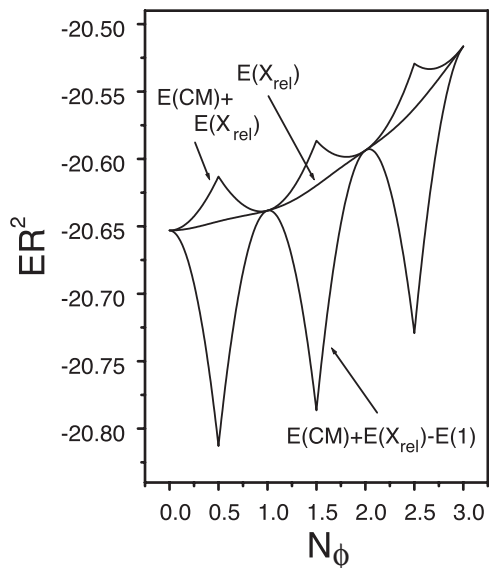


Fig. 4. Calculated emission energies for the X^- complex as a function of the number of flux quanta. We show the energy of the relative motion $E(X_{\text{rel}})$, the total energy of the complex $E(\text{CM}) + E(X_{\text{rel}})$, and the difference between this energy and the final state after recombination (single electron) $\omega = E(\text{CM}) + E(X_{\text{rel}}) - E(1)$

Conclusion In conclusion, we analysed the behaviour of a negatively charged exciton X^- on a ring with infinitesimal thickness in an external magnetic field. The total X^- Hamiltonian can be separated into the center of mass and relative parts. The magnetic field dependence in both parts enters only into the kinetic energy operators. Therefore we first consider the noninteracting system. In this case the single particle ground state energy exhibits oscillations as a function of the number N_ϕ of flux quanta threading the ring. The sharp maxima, seen at half-integer N_ϕ , mark changes of the ground state angular momentum. For interacting particles we find that in the ring geometry the ratio of Coulomb to kinetic energies is proportional to the ring radius, so that for the radius large enough the interaction effects dominate. Therefore an exciton localised on a ring with a small enough radius will behave like a pair of weakly interacting particles, with oscillations of the ground state energy, similar to the single-particle case. In contrast, for large radii the exciton and the hole move collectively, and the exciton energy depends smoothly upon the magnetic field, whose influence is reduced to the diamagnetic shift. In the case of the X^- complex the total ground state energy contains two components: (i) the center-of-mass component, behaving like a heavy charged single particle, and thus oscillating with the magnetic field, and (ii) the relative component, carrying all interactions and exhibiting single-particle-like oscillations for small ring radii, and the featureless diamagnetic shift for large radii. In a magneto-PL experiment we probe the difference between this total energy and the energy of the final state after recombination (a single electron). Due to the large mass of the X^- complex, this photon energy is dominated by the final state contribution, and shows oscillations for all ring radii.

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