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Brief Reports

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Delocalization of excitons in a magnetic field

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The two-body effects for a Mott-Wannier exciton in a magnetic field are investigated. For excitons with nonvanishing pseudomomentum there exists an outer potential well which contains a class of weakly bound and delocalized quantum states. The spectrum and eigenfunctions in this potential well are calculated in a harmonic approximation.

The behavior of matter in strong magnetic fields became in the past two decades a subject of great interest. The numerous investigations in different branches of physics like, for example, atomic, molecular, and solid-state physics, have shown that the properties of matter in strong external fields are very different from those of the field-free case and we, therefore, encounter a variety of new phenomena due to the presence of the strong field. The term "strong field" has no absolute meaning but rather indicates that the forces due to the field are comparable to or even larger than the interaction forces of the system. This can happen for different states of the same physical system on different scales of the absolute field strength. In particular, it is possible to investigate the strong field regime of, for example, the hydrogen atom by studying its highly excited Rydberg states at laboratory magnetic-field strengths.[1] In solid-state physics, the analog of such a "simple" system like the hydrogen atom would be an elementary excitation like, for example, an exciton. Let us assume that the distance of the particle and hole is much larger than the lattice spacing, i.e., we consider a Mott-Wannier exciton. For this kind of exciton all effects due to the presence of the lattice can be included in a simple way in the excitonic Hamiltonian model.[2,3] This Hamiltonian model is a phenomenological approach which neglects many residual interactions.[2] However, it represents under certain circumstances a good zeroth-order approximation to the exact excitonic Hamiltonian.

Excitons, excitonic molecules, as well as excitonic matter, have been studied extensively in the literature.[2–5] They are common phenomena occuring in insulator and semiconductor physics. Since the dielectricity constant can become large and since the effective masses of the particle and hole are very often small compared to the proton and electron mass, the corresponding excitonic Rydberg (binding energy of the exciton) can be much smaller than the hydrogenic one. This has drastic consequences if we turn on a magnetic field: the strong-field regime occurs for the ground or first few excited states of certain excitons already at laboratory magnetic-field strengths. They are, therefore, ideal objects in order to study strong-field effects in the laboratory (see, for example, Refs. 2, 3, or 6–8). For an interacting two-body system, like an exciton, in a homogeneous magnetic field it is not possible to perform a complete separation of the center-of-mass (CM) motion in the corresponding Hamiltonian. In other words, it is not possible to completely decouple the center-of-mass and internal motion. Since the masses of the particle and hole are, in general, of comparable order of magnitude, all two-body effects are of particular importance and should be treated exactly in the underlying excitonic Hamiltonian. A study of these two-body effects is precisely the subject of the present paper. (We remark that both the coupling of the collective to the relative motion as well as the two-body effects for the internal relative motion have been investigated very recently[9–12] for the case of the hydrogen atom in a magnetic field.)

Our starting point is the nonrelativistic Hamiltonian for the neutral exciton in a uniform magnetic field. We assume that the pseudoseparation of the center-of-mass motion has been performed (for the details of this well-known transformation we refer the reader to the literature[13–17]). The resulting Hamiltonian takes on the following appearance:

$$H=\frac{1}{2M}(\mathbf{K}-e\mathbf{B}\times\mathbf{r})^2+\frac{1}{2\mu}\left[\mathbf{p}-e\frac{\mu}{\tilde{\mu}}\mathbf{A}\right]^2-\frac{e^2}{\epsilon|\mathbf{r}|}\ , \qquad (1)$$

where $\tilde{\mu}=[m_p m_h/(m_h-m_p)]$ (we assume $m_h>m_p$). m_h and m_p are the mass of the hole and particle, respectively. $\mu=(m_p m_h/M)$, M, $\mathbf{B}$, and ϵ are the reduced and total mass, magnetic field vector, and dielectricity constant, re-

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spectively. **K** is the pseudomomentum. **A** is the vector potential which is in the following chosen to be in the symmetric gauge $\mathbf{A}(\mathbf{r})=(1/2)\mathbf{B}\times\mathbf{r}$. **r** is the relative coordinate of the particle and hole and **p** its canonical conjugated momentum. The model Hamiltonian (1) for the excitation is based on two approximations: the effective-mass approximation and the dielectric continuum model for Coulomb screening. These approximations can hold only if the radius of the exciton is much larger than the lattice spacings of the solid. The canonical conjugated coordinate belonging to the pseudomomentum **K** is the center-of-mass coordinate **R**. Since **K** is a constant of motion, the center-of-mass coordinate is a cyclic coordinate and does not appear in the transformed Hamiltonian (1). However, this does not mean that the center-of-mass motion decouples from the internal motion. Indeed the equation of motion for the center of mass resulting from the Hamiltonian (1) reads as follows:

$$\dot{\mathbf{R}}=\frac{1}{M}\mathbf{K}-\frac{e}{M}\mathbf{B}\times\mathbf{r}\ , \qquad (2)$$

i.e., the center-of-mass velocity is, apart from a constant, completely determined by the internal relative coordinate. From Eq. (2) it is clear that the center of mass is, even for the case of vanishing pseudomomentum, intimately coupled to the internal motion. Let us investigate the classical CM motion of the exciton for the special case of vanishing pseudomomentum $\mathbf{K}=\mathbf{0}$ and for varying total energy E. The Hamiltonian (1) possesses for $\mathbf{K}=\mathbf{0}$ an additional internal rotational symmetry around the magnetic-field axis (which is assumed to be oriented along the z axis) and the projection L_z of the internal angular momentum onto the magnetic-field axis is, therefore, a conserved quantity. For simplicity we consider the case $L_z=0$. The resulting Hamiltonian H_0 shows, with increasing energy, a transition from regularity to chaos for the internal motion $[\mathbf{r}(t),\mathbf{p}(t)]$. It is now interesting to ask the following: what does happen with the CM motion of the exciton if its internal motion passes from regularity to chaos?

For regular internal motion the CM trajectories perform in the plane perpendicular to the magnetic field (note: the CM motion parallel to the field axis is free) quasiperiodic oscillations with small amplitude which are confined to a bounded range of coordinate space. If we increase the total energy, chaotic trajectories for the internal motion appear and take over more and more of phase space. Eventually, complete phase space becomes chaotic. For chaotic internal motion, the CM motion is no longer restricted to some bounded volume of phase space but experiences with increasing time an increasing volume of the two-dimensional coordinate space. A second important observation is that the CM motion closely resembles a random motion which has its origin in the intrinsic chaotic force $-e(\mathbf{B}\times\dot{\mathbf{r}})$ [see Eq. (2)]. A characteristic feature of a random motion is its diffusion law, i.e., the time dependency of the traveled mean-square distance. In order to investigate this property for our case of the excitonic CM motion (for the case of chaotic integral motion) we have calculated the mean-square distance $\langle X^2+Y^2\rangle$ as a function of time for an ensemble of 500 CM trajectories. The result is presented in Fig. 1. Within statistical accuracy, the plot shows a linear dependency $\langle X^2+Y^2\rangle=Dt$. D is the diffusion constant for the spreading of the ensemble of CM trajectories and has, in our case, an approximate value of $D=5.75\times10^5$ Å^2/ns. To summarize, we have shown that the classical transition from regularity to chaos in the internal relative degrees of freedom is accompanied by a transition from localized bounded quasiperiodic to delocalized unbounded randomlike motion in the center of mass of the exciton. The randomlike CM motion of the exciton has diffusive properties in the sense that it obeys a linear diffusion law for the spreading in a plane perpendicular to the magnetic field. This might have implications on those properties of solids which have a contribution from excitonic processes like, for example, the electro(optical) properties of semiconductors in a magnetic field. However, there are two points which should be kept in mind and which deserve further investigation. The first point is the fact that we have studied the classical CM motion of the exciton and it is not obviously what will happen if we quantize both the internal as well as the CM motion. Does the CM diffusion survive or does quantum localization take place? The second point is that we have chosen a zeroth-order model Hamiltonian for the Mott-Wannier exciton in a magnetic field which neglects many residual interactions and one may ask how these interactions perturb and modify the excitonic motion in the magnetic field. Answers to these questions would help to clarify the role of the excitonic CM motion for the properties of solids. However, they go beyond the scope of this paper and are left to future investigations.

Next let us investigate the influence of the two-body character on the behavior and properties of the exciton with nonvanishing pseudomomentum. Our starting point is again the Hamiltonian (1) for $\mathbf{K}\neq\mathbf{0}$. The first quadratic term of this Hamiltonian represents the kinetic energy

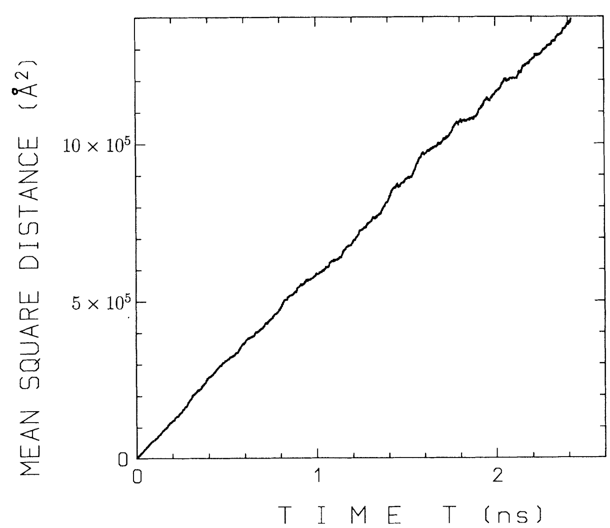

FIG. 1. The mean-square distance $\langle X^2+Y^2\rangle$ of an ensemble of 500 center-of-mass trajectories in the fully chaotic regime as a function of time. The parameter values are $m_p=0.2m_e$, $m_h=5m_e$, $\epsilon=5$, $B=2.35$ T, and a total energy of $E\approx-0.75$ meV.

of the CM motion $(M/2)\dot{\mathbf{R}}^2$ [see Eq. (2)] whereas the second quadratic term is the internal kinetic energy $(\mu/2)\dot{\mathbf{r}}^2$. The important observation is now that *each term of the quadratic kinetic energy of the center of mass* $(1/2M)(\mathbf{K}-e\mathbf{B}\times\mathbf{r})^2$ *is gauge invariant and acts as a potential energy for the internal relative motion* (see Ref. 12 for an explanation). From the point of view of the internal motion the Hamiltonian (1) can, therefore, be divided in a unique way in kinetic (T) and potential (V) energy terms, i.e., $H=T+V$ where

$$T=\frac{1}{2\mu}\left[\mathbf{p}-\frac{e}{2}\frac{\mu}{\tilde{\mu}}\mathbf{B}\times\mathbf{r}\right]^2 \tag{3a}$$

and

$$V=\frac{1}{2M}(\mathbf{K}-e\mathbf{B}\times\mathbf{r})^2-\frac{e^2}{\epsilon|\mathbf{r}|} . \tag{3b}$$

In the following, we discuss the properties of the potential V and the resulting consequences for the excitonic dynamics. Apart from the constant $\mathbf{K}^2/2M$, the potential V is a combination of a diamagnetic $(e^2/2M)(\mathbf{B}\times\mathbf{r})^2$, a Stark $(e/M)(\mathbf{B}\times\mathbf{K})\mathbf{r}$, and a Coulomb potential term $(-e^2/\epsilon|\mathbf{r}|)$. With the choices $\mathbf{B}=(0,0,B)$ and $\mathbf{K}=(0,K,0)$ $(K>0)$ the electric field points along the negative x direction. Figure 2 shows an intersection of the potential V along the direction of the electric field ($y=z=0$). Close to the origin, i.e., for small absolute values of the x coordinate, the Coulomb potential dominates. With increasing absolute values of the x coordinate, the Stark term gains in significance and eventually becomes comparable with the strength of the Coulomb potential. The diamagnetic term provides in this coordinate region only a small correction to the Coulomb and Stark terms. As a consequence of the competition of the latter two terms, a saddle point arises which is, in Fig. 2, located at approximately $x\approx-807$ Å. For even larger absolute values of the x coordinate the Coulomb potential becomes smaller and the shape of the potential is more

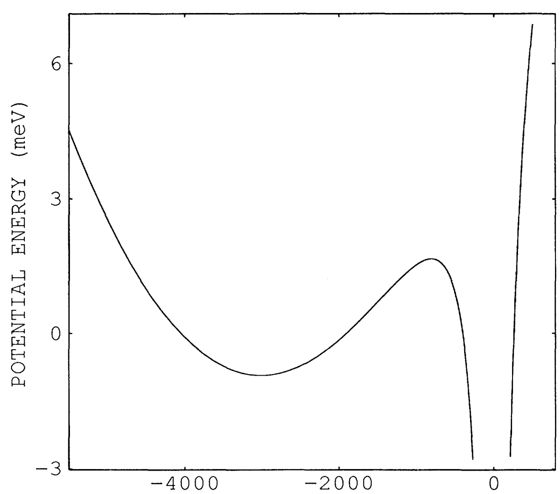

FIG. 2. The potential V along the direction (x axis) of the motional electric field. The parameter values are the same as in Fig. 1 and in addition we have $K=1.3\times10^6 m_e$ Å/ns.

and more determined by the quadratic potential term $(e^2/2M)B^2x^2$. Due to the competition of the Stark and diamagnetic terms the potential V now develops a minimum which is, in Fig. 2, located at approximately $x\approx-3004$ Å. Not only the quantitative appearance of the potential V depends, of course, on the values of M,ϵ,K,B but also the existence of both the saddle point as well as the minimum. Quantitative conditions for their existence will be given below.

The discussed properties of our potential V have important implications on the dynamical behavior of the exciton. One observation is that the ionization of the exciton, i.e., the infinite separation of the particle and hole can take place only in the direction parallel to the magnetic field: in the direction perpendicular to the magnetic field, the diamagnetic potential term is dominating for large distances and causes a confining behavior of the potential V. Another important observation is the fact that the minimum of the potential V leads to a potential well and, as we shall see below, to a new class of weakly bound quantum states inside this well. Excitons in these states are very extended delocalized objects for which the particle and hole are separated far from each other.

Let us turn now to a quantitative investigation of the potential V. The outer potential well described above and illustrated in the intersection of Fig. 2 exists only if the potential V possesses both a saddle point and a minimum. In order to obtain a condition for their existence we have to determine the extrema of the potential V. A simple calculation shows that the components of the coordinate vectors $\{\mathbf{r}_0\}$ of the extrema obey the equations $y_0=z_0=0$ and

$$x_0^3+(K/B)x_0^2-(M/\epsilon B^2)=0 . \tag{4}$$

The existence of both the saddle point and the minimum yield the following condition on the parameter values:

$$K^3>\tfrac{27}{4}(BM/\epsilon) . \tag{5}$$

This is a necessary and sufficient condition which says that the pseudomomentum must exceed some critical value in order to form the outer potential well. In the following, we assume that the condition (5) is fulfilled. The question arises then: does there exist bound states in the outer potential well and how do they look?

The simplest way to investigate this question is to expand the potential V around its minimum position $\mathbf{r}_0$ up to second order and to solve the resulting equations of motion analytically. The Taylor expansion of V around $\mathbf{r}_0$ yields the following approximate potential:

$$V_a=C+(\mu/2)\omega_x^2x^2+(\mu/2)\omega_y^2y^2+(\mu/2)\omega_z^2z^2 , \tag{6}$$

where

$$\omega_x=\left[\frac{2}{\mu}\left[\frac{B^2}{2M}+\frac{1}{\epsilon x_0^3}\right]\right]^{1/2} ,$$

$$\omega_y=\left[\frac{1}{\mu}\left[\frac{B^2}{M}-\frac{1}{\epsilon x_0^3}\right]\right]^{1/2} ,$$

$$\omega_z=\left[\frac{1}{\mu}\left[\frac{1}{\epsilon|x_0|^3}\right]\right]^{1/2} ,$$

$C=(K^2/2M)+(2/\epsilon x_0)-(B^2/2M)x_0^2$ is a constant. V_a is the potential for a three-dimensional anisotropic harmonic oscillator. The approximate Hamiltonian describing the motion inside the potential well has the form $H_a=(T+V_a)$ where T is the kinetic energy for a particle with charge $e(\mu/\tilde{\mu})$ and mass μ in a magnetic field. In the following we investigate the classical trajectories as well as the quantum-mechanical spectrum of the Hamiltonian H_a. To this end we solve the gauge-invariant Newtonian equations of motion belonging to the Hamiltonian H_a by the ansatz

$$\begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} a \\ b \end{pmatrix} \exp(i\omega t) , \quad z=c\exp(i\omega_z t) , \tag{7}$$

which yields the following solutions for the normal modes in the (x,y) plane:

$$\omega_\pm=\frac{1}{\sqrt{2}}\{(\omega_x^2+\omega_y^2+\omega_c^2)\pm[(\omega_x^2+\omega_y^2+\omega_c^2)^2 -4\omega_x^2\omega_y^2]^{1/2}\}^{1/2} , \tag{8}$$

where $\omega_c=(B/\tilde{\mu})$ is the cyclotron frequency. We obtain two normal frequencies $\omega_\pm$ for the oscillatory motion perpendicular to the magnetic field and one frequency ω_z for the harmonic motion parallel to the magnetic field. The quantum-mechanical spectrum belonging to the above classical trajectories is a harmonic-oscillator spectrum and the binding energies of the corresponding states read as follows:

$$E=C_0-(n_+ +\tfrac{1}{2})\omega_+ -(n_- +\tfrac{1}{2})\omega_- -(n_z+\tfrac{1}{2})\omega_z , \tag{9}$$

where $C_0=(B/2\mu)-C$ and $(B/2\mu)$ is the zero point energy of the free particle and hole in a magnetic field, i.e., the ionization threshold.

For our example in Fig. 2, the binding energy of the ground state inside the potential well is approximately $E_B\approx 0.9$ meV. The order of magnitude of the spatial extension of the ground state is roughly 300 Å in both the x and y direction and about 1100 Å in the z direction (for comparison, the corresponding exciton in field-free space has a binding energy of 105 meV and a Bohr radius of 14 Å). The energies associated with the frequencies ω_+, ω_-, and ω_z take on the values $E_+=1.36$ meV, $E_-=0.053$ meV, and $E_z=0.065$ meV. Since the threshold energy is approximately 0.71 meV we have a large number of bound states in the potential well. However, only the ground and first few excited states can, for our example of the parameter values, be described accurately within the harmonic approximation. For large amplitudes inside the well, i.e., higher excitations, anharmonicity corrections become important and our harmonic-oscillator expansion breaks down. The range of validity of the harmonic-oscillator approach depends sensitively on the parameter values, i.e., on the masses, dielectricity constant, and field strength. For certain excitons, anharmonicity corrections are important already for the ground state in the potential well. We remark that the size of the excitonic states varies strongly for different parameter values.

For the delocalized quantum states discussed above, the particle and hole are always separated far from each other and are prevented from coming close together by a potential barrier. In reality, these quantum states possess a finite lifetime due to the possibility of the tunneling process to the Coulomb singularity well. However, in the bulk the potential barrier might prevent the particle and hole from recombination and might increase the lifetime of the exciton. It is now a challenging question to ask whether the two phenomena described in this paper might be observable in direct experiments or might contribute to certain properties of solids.

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