Giant-dipole excitons in cuprous oxide

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Abstract. In this work we present the interaction potentials and eigenenergies of a novel species of Wannier excitons when exposed to crossed electric and magnetic fields. More precisely, we present the theory of giant-dipole excitons in Cu$_2$O. We derive an exact formulation of the field-dressed excitonic Hamiltonian and exemplarily calculate the excitonic eigenenergies in an exact diagonalization approach for external field strengths of $B = 4$ T and $E = 1$ kV/cm. For this particular field configuration, we obtain level spacings between $1.14 \mu$eV and $77.6 \mu$eV.

1. Introduction
In semiconductor physics, a bound state between an excited electron in the conduction band and a positively charged hole in the valence band is known as exciton. As the interaction between both constituents can be modeled by a screened Coulomb interaction, an exciton is often considered to be a solid state analogue to the hydrogen atom [1, 2]. For instance, recent measurements of highly excited Rydberg excitons up to principal quantum numbers $n = 25$ have attracted quite some attentions [3]. However, the simple hydrogen model is incapable in reproducing the experimentally observed level-splitting due to fine- and hyperfine splitting, thus it has been expanded by considering both the complex level structure and the cubic symmetry $O_h$ of Cu$_2$O [4]. In the presence of external electric and magnetic fields the symmetry of the excitonic states is further reduced, leading to level structure providing numerous complex splittings of excitonic absorption lines [5, 6].

In atomic physics, an exotic state of highly excited Rydberg states in crossed external fields are so-called giant-dipole states [7]. When the separation of relative and center-of-mass degrees of freedom are treated correctly, the effect of the center-of-mass motion on the internal degrees of freedom is an effective potential that gives rise to an outer potential well for certain applied field strengths. Bound states in these outer potential wells are characterized be large spatial electron-hole separations up to several micrometers, leading to huge permanent electric dipole moments. However, the concept of giant-dipole states is not only restricted to atomic systems but, in principle, to any neutral bound quasi-particle species such as excitons as well. In a recently published work [8], a sufficient theoretical description of excitons exposed to external fields in Cu$_2$O has been derived. In this work the exact separation between gauge-dependent and gauge-independent terms was performed, giving the possibility of identifying both the kinetic as well as the exact single-particle potential for internal degrees of freedom.

In this paper, we present a brief derivation of the irreducible tensor representation of the excitonic Hamiltonian in external electric and magnetic fields. Following this, we derive giant-dipole potential surfaces for various external field strengths. Furthermore, within an exact diagonalization approach we calculate excitonic eigenspectra [9].
2. The excitonic Hamiltonian in external fields

We consider excitons in Cu$_2$O which are formed by an electron in the energetically lowest $\Gamma_0^+$-conduction band and a positively charged hole in the uppermost $\Gamma_0^+$-valence band, where the latter is triply degenerate. These three bands are deformed due to interband interactions and non-spherical symmetry properties of the solid. However, an effective description can be achieved by an $I = 1$ quasi-spin representation in the hole degrees of freedom [10].

The hole Hamiltonian is characterized by three Luttinger parameters $\gamma_i$, $i=1,2,3$ [11]. The mapping $\{ab\} \equiv (ab + ba)/2$ is called the symmetric product and $c.p.$ denotes the cyclic permutations of terms. Furthermore, the quasi-spin $I$ couples to the hole-spin $S_h$ via

$$H_{so} = \frac{2}{3} \Delta (1 + S_h \cdot I),$$

while the electron spin $S_e$ is not considered in this work.

In the case that an external magnetic field is applied, the canonical momenta $p_{e,h}$ are replaced by $p_{e,h} \to p_{e,h} \pm A(r_{e,h})$, where $A(r)$ is the magnetic vector potential. The coupling of $S_h$ and $I$ to the external magnetic field is given by

$$H_B = \mu_B [(3\kappa + \frac{gs}{2})I \cdot B - gsS_h \cdot B].$$

If an external homogeneous electric field is applied, the stark terms $\mp E \cdot r_{e,h}$ have to be added to the Hamiltonian. If one introduces the relative coordinate $r = r_e - r_h$ and center-of-mass vector $R$, one can perform a pseudoseparation of both degrees of freedom using the eigenstates and eigenvalues $K$ of the so-called pseudomomentum $\tilde{K} = P - (B \times r)/2$ [7]. As it has been shown in previous studies, the excitonic Hamiltonian can then be transformed into a single-particle Hamiltonian $H_{ex} = T + V_{gd}$ with

$$T = H_e(\pi) + H_h(\pi), \quad \pi_i = p_i - qA_i(r) - \sum_k \left( \frac{m_i}{M} \delta_{ki} - \Omega_i k \right) \tilde{K}_k, \quad I_{ij} = 3\{I_i I_j\} - 2\delta_{ij},$$

$$V_{gd}(r) = \left( \Omega_1 \tilde{K}^2 + E \cdot r - \frac{1}{r} \right) I_1 - \Omega_2 \sum_i \tilde{K}_i^2 I_{ii} - \frac{2}{3} \Omega_3 \sum_{i,j<i} \tilde{K}_i \tilde{K}_j I_{ij} + H_{so} + H_B,$$

$q = (m_e - m_h)/M$, $M = m_e + m_h$ and $\tilde{K} = K + B \times r$ [8]. Here, the term $V_{gd}$ represents a single-particle potential for the internal motion of the field-dressed excitons.

3. Excitonic giant-dipole potential surfaces and eigenenergies

Due to the spin-1 and spin-1/2 degrees of freedom, the potential $V_{gd}$ can be expressed as a $6 \times 6$ matrix, where the matrix elements are functions of the external field parameters $B || [100]$, $E || [001]$ and the spatial coordinate $r$, respectively. The eigenvalues $V_i(r)$, $i = 1, \ldots, 6$ are three-dimensional potential surfaces which parametrically depend on the external field strengths.
In Fig. 1(a) we show a one dimensional cut through the six potential curves along the negative $z$-direction for applied field strengths of $B = 4$ T and $E = 1$ kV/cm. The main figure shows the potential curves $V_3(z)$ (green solid curve) and $V_5(z)$ (red solid curve), respectively. Obviously, both potential curves possess local minima at $z \approx -0.2 \mu$m and $z \approx -0.16 \mu$m. This large electron-hole separation results in a huge electric dipole moment, justifying the expression excitonic giant-dipole states. The potential curves $V_4(z)$ and $V_6(z)$ are not shown in the main figure as they are not resolved on this energy scale. However, in the inset we present both $V_1(z)$ (solid blue line) and the adjacent curve $V_2(z)$ (dashed blue line). As one can see, these two curves possess local minima at $z \approx -0.1775 \mu$m with an energetic separation of around $3 \mu$eV.

In Fig. 1(b) the two-dimensional potential surface $V_1(x,z)$ is shown for fixed $y = 0$. In this plot one clearly sees the local potential minimum at $x = 0, z = -0.1775 \mu$m.

Figure 2. Excitonic eigenenergies for $B = 4$ T and $E = 1$ kV/cm. We find four bound states with an energetic separation between 1.14 $\mu$eV and 77.6 $\mu$eV.

For the calculation of the excitonic eigenenergies, we apply an exact diagonalization approach. In particular, in case the couplings among the single potential surfaces are neglected, one can
derive analytic solutions for the bound states in the outer potential wells. Together with the basis function $|1, m\rangle$, $m = -1, 0, 1$ and $|1/2, \pm 1/2\rangle$ for the spin-1 and spin-1/2 Hilbert spaces, these functions can be used to define a basis set for the exact diagonalization of the excitonic Hamiltonian given by Eq. (4). As we are mostly interested in the energetically low lying states, we have used the giant-dipole functions of the potential surface $V_1$ for the exact diagonalization.

In Fig. 2 we present the excitonic eigenenergies for applied field strengths of $B = 4$ T and $E = 1$ kV/cm. For comparison, we show a one-dimensional cut through the energetically lowest potential surface $V_1$ as well. For this surface we also indicate the ionization limit, i.e. the energy above which all excitonic states lie in the continuum. For the applied field configuration this energy is given by $-9.43$ meV, whereby the ground state energy is given by $-9.58$ meV. In total, we find four bound excitonic giant-dipole states. Their energetic spacing is determined by two energy scales. The first one is given by $1.14$ µeV which is the spacing between the ground state and the first excited state as well as between the second and third excitation. This spacing is not resolved in the main figure but presented in the inset where we indicate the energetic separation between the ground state and the first excited state. The second energy scale is given by $77.6$ µeV and is much larger than the first one. This is the spacing between the first and second excited state and is clearly visible in Fig. 2. As all bound states are quite close to the ionization limit, no more bound states are found for this particular field configuration.

In general, we find that bound excitonic states are only found for sufficiently strong magnetic fields as in this case the outer potential minima are deep enough to provide bound states. For instance, for $E = 1$ kV/cm giant-dipole excitons are only found for $3.4$ T $\leq B \leq 4.4$ T where we find between one and six bound states with energetic level spacings in the range of $0.6 ... 100$ µeV.

4. Summary and conclusions
In the present article, we have presented a theoretical description of field-dressed excitons in Cu$_2$O. In particular, we have shown the exact kinetic and potential energy terms which can be derived from a pseudoseparation of the relative and center-of-mass degrees of freedom. We obtain potential energy surfaces for the internal motion, possessing local minima providing bound excitonic giant-dipole states. In future studies one might address the experimental preparation and verification of excitonic giant-dipole states. One possible approach may be to use time-dependent external fields to apply an adiabatic state transfer of highly excited Rydberg excitons into giant-dipole states. The experimental verification could be achieved via microwave spectroscopy and via the direct measurement of the large electric dipole moment.

Acknowledgments
We acknowledge support by the Focus Programme SPP 1929 GiRyd by the Deutsche Forschungsgemeinschaft (DFG).

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