Magneto-Stark-effect of yellow excitons in cuprous oxide

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We investigate and compare experimental and numerical excitonic spectra of the yellow series in cuprous oxide Cu₂O in the Voigt configuration and thus partially extend the results from Schweiner et al. [Phys. Rev. B 95, 035202 (2017)], who only considered the Faraday configuration. The main difference between the configurations is given by an additional effective electric field in the Voigt configuration, caused by the motion of the exciton through the magnetic field. This Magneto-Stark effect was already postulated by Gross et al. and Thomas et al. in 1961 [Sov. Phys. Solid State 3, 221 (1961); Phys. Rev. 124, 657 (1961)]. Group theoretical considerations show that the field most of all significantly increases the number of allowed lines by decreasing the symmetry of the system. This conclusion is supported by both the experimental and numerical data.

I. INTRODUCTION

Application of electric or magnetic fields, both representing well-controlled external perturbations, has offered detailed insight into quantum mechanical systems by inducing characteristic shifts and lifting degeneracies of energy levels. This potential became apparent already for the simplest quantum mechanical system in nature, the hydrogen atom, for which an external field reduces the symmetry from spherical to cylindrical [1–3].

In condensed matter, the hydrogen model is often applied as a simple, but nevertheless successful model for problems in which Coulomb interaction mediates the coupling between opposite charges of quasi-particles [4, 5]. The most prominent example is the exciton, the bound complex of a negatively charged electron and a positively charged hole. In particular, for crystals of high symmetry such as cubic systems, the phenomenology of energy levels in an external field often resembles the corresponding hydrogen spectra after rescaling the energy axis with the Rydberg energy such as the effective mass and the dielectric constant which in these systems are isotropic.

Recent high resolution spectroscopy of excited exciton states in Cu₂O has allowed one to reveal features which result from the crystal environment having discrete instead of continuous symmetry, leading to deviations from a simple hydrogen description [6, 7]. For example, the level degeneracy is lifted already at zero field. A detailed analysis shows that this splitting arises mostly from the complex valence band structure [8–10]. In effect, angular momentum is not a good quantum number anymore, but states of different angular momenta having the same parity are mixed. As the exciton size is large compared to that of the crystal unit cell, the mixing is weak, however, so that angular momentum may still be used approximately as quantum number. Simply speaking, this can be understood in the following way: The exciton wave function averages over the crystal lattice, assembled by the cubic unit cells. With increasing wave function extension a state becomes less and less sensitive to the arrangement of the atoms in a crystal unit cell and thus to the deviations from spherical symmetry.

For example, the size of the cubic unit cell in Cu₂O is about 0.4 nm [11], while the Bohr radius is 1.1 nm for the dipole-active P-type excitons dominating the absorption [12]. The size of the lowest exciton \( n = 4 \), for which the mixing becomes optically accessible, is 25.5 nm, covering thousands of unit cells. As a consequence of the weak mixing, the state splitting is small compared to the separation between levels of different \( n \). Generally, the average radius of the wavefunction increases as \( n^2 \), so that the zero-field state splitting decreases with increasing principal quantum number, leading to the levels becoming quasi-degenerate again for \( n \) larger than about 10, as demonstrated in Ref. [7]. For smaller principal quantum numbers, the splitting can be well resolved, in particular, because the mixing of levels also causes to a redistribution of oscillator strength, so that not only the \( P \)-states but also higher lying odd states such as \( F, H, \ldots \) within a multiplett can be resolved in single photon absorption.

Applying a magnetic field leads to a squeezing of the wave function mostly normal to the field, so that the influence of the crystal is enhanced. Even when assuming spatial isotropy, the symmetry is reduced to rotational invariance about the field, leaving only the magnetic quantum number \( m \) as conserved quantity. As a consequence, states of the same \( m \) but different orbital angular momentum become mixed, activating further lines optically. In combination with the Zeeman splitting of the levels this leads to a rich appearance of the absorption spectra, in particular, as for excitons due to their renormalized Rydberg energy being much smaller than in the atomic

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case, also resonances between states of different principal quantum numbers can be induced by the field application, at least for excited excitons.

As indicated above, a key ingredient for the renormalized Rydberg energy are the smaller masses of the involved particles forming an exciton, in particular of the hole compared to the nucleus. In Cu$_2$O, the electron mass is almost that of the free electron ($m_e = 0.99m_0$ [13]), while the hole represents the lighter particle ($m_h = 0.58m_0$ [13]), in contrast to most other semiconductors. This makes the reduced mass entering the relative motion of electron and hole only a factor of less than 3 smaller than in hydrogen. The mass of the exciton center of mass motion, on the other hand, is clearly more than three orders of magnitude smaller. This difference raises the question whether this mass difference causes a difference of the optical spectra in magnetic field, as the excitons are generated with a finite wavevector identical to the wavevector of the exciting laser.

In that respect two different field configurations need to be distinguished, namely the Faraday and the Voigt configuration. In the first case the magnetic field is applied along the optical axis, while in the second case the field is oriented normal to the optical axis. Using, for example, exciting light with polarization normal to the light wavevector, one would not expect a difference in the spectra for atoms.

Here we have performed corresponding experiments which demonstrate that in contrast to the atomic case the exciton spectra differ significantly for the two field configurations. These findings are in good agreement with detailed theoretical calculations. From the comparison we trace the difference to the the Magneto-Stark-effect [14, 15] which is acting only in the Voigt configuration, where the excited exciton is moving normal to the magnetic field, so that its two constituents are subject to the Lorentz force acting in opposite direction for electron and hole and therefore trying to move them apart, similar to the action of an electric field of the form

$$F_{\text{MSE}} = \frac{\hbar}{M} (K \times B),$$

where $M$ is the exciton center-of-mass, $K$ is the exciton wavevector, and $B$ is the magnetic field. The action of this field is obviously absent in the Faraday configuration.

**II. HAMILTONIAN**

Our theoretical description of excitonic spectra in Cu$_2$O with an external magnetic field builds upon Schweiner et al.’s treatment in Ref. [9], where only the Faraday configuration was considered. The Hamiltonian without magnetic field is given by

$$H = E_g + H_e(p_e) + H_h(p_h) + V(r_e - r_h),$$

where $H_e$ and $H_h$ are the kinetic energies of the electron and hole, respectively. They are given by

$$H_e(p_e) = \frac{p_e^2}{2m_e}$$

and

$$H_h(p_h) = H_{so} + \left\{ h^2 \left( \gamma_1 + 4\gamma_2 \right) p_h^2 \right. + 2 (\eta_1 + 2\eta_2) p_h^2 (I \cdot S_h)$$

$$- 6\gamma_2 (p_h^2 I_1^2 + \text{c.p.}) - 12\eta_2 (p_h^2 I_1 S_h_1 + \text{c.p.})$$

$$- 12\gamma_3 (\{p_h_1, p_h_2\} \{I_1, I_2\} + \text{c.p.})$$

$$- 12\eta_3 (\{p_h_1, p_h_2\} (I_1 S_h_2 + I_2 S_h_1) + \text{c.p.}) \right\}$$

with the spin-orbit interaction

$$H_{so} = \frac{2}{3} \Delta \left( 1 + \frac{1}{\hbar^2} I \cdot S_h \right).$$

Here, $I$ is the quasi-spin and $S_h$ the spin $S_h = \frac{1}{2}$ of the hole and c.p. denotes cyclic permutation. Electron and hole interact via the screened Coulomb potential

$$V(r_e - r_h) = \frac{e^2}{4\pi\varepsilon_0\varepsilon |r_e - r_h|},$$

with the dielectric constant $\varepsilon$. To account for the magnetic field $B$, we use the minimal substitution $p_e \rightarrow p_e + eA(r_e)$ and $p_h \rightarrow p_h - eA(r_h)$ with the vector potential for a homogenous field $A(r_{e,h}) = (B \times r_{e,h})/2$. The energy gained by the electron and hole spin in the external magnetic field is described by

$$H_B = \mu_B [(g_e S_e + (3\kappa + g_s/2)) I - g_s S_h] \cdot B/\hbar,$$

with the Bohr magneton $\mu_B$ and the $g$-factor of the hole spin $g_s \approx 2$. We finally switch into the center of mass reference frame [16]:

$$r = r_e - r_h,$$

$$R = m_e + m_h r_e + m_e + m_h r_h,$$

$$p = \hbar k - eB \times R = \frac{m_h}{m_e + m_h} p_e - \frac{m_e}{m_e + m_h} p_h,$$

$$P = \hbar K + eB \times r = p_e + p_h,$$

and set $K = 0$ and $R = 0$ for now. More details can be found in Refs. [9, 17–19] and values of material parameters for Cu$_2$O used in Eqs. (2)-(8) are listed in Table I.

**A. Faraday and Voigt configuration, Magneto-Stark-effect**

We consider two different relative orientations of the magnetic field to the optical axis. In the Faraday configuration, both axes are aligned to be parallel, whereas
in the Voigt configuration, they are orthogonal to each other. Generally, the exciting laser will transfer a finite momentum $\hbar \mathbf{K}$ onto the exciton. This center of mass momentum would have to be added in the terms for the kinetic energies. Even without a magnetic field, this leads to quite complicated formulas (cf. the expressions for the Hamiltonian in the supplemental material of Ref. [22]) which are further complicated by the minimal substitution. Since the effect of many of the arising terms is presumably negligible due to the smallness of $\mathbf{K}$, we simplify the problem and only consider the leading term [3, 23] for the central cell corrections with the Haken potential as given in Refs. [18, 22] in our treatment to correctly take the coupling to the low lying S states into account.

### III. NUMERICAL APPROACH

Using the Hamiltonian (2) with the additional terms for the central cell corrections $H_{\text{ccc}}$ and the Magneto-Stark effect $H_{\text{ms}}$ with a suitable set of basis vectors, the Schrödinger equation can be brought into the form of a generalized eigenvalue equation

$$
D \psi = E M \psi
$$

We choose a basis consisting of Coulomb-Sturmian functions with an appropriate part for the various appearing spins and angular momenta. Due to the broken inversion symmetry, it is not sufficient to include only basis functions of odd parity as in reference [9]. Instead, basis functions of even symmetry have to be included as well. The resulting equation can then be solved using a suitable LAPACK routine [24]. For details we refer to the discussions in Refs. [9, 18, 25].

#### A. Oscillator strengths

The extraction of the dipole oscillator strengths is performed analogously to the calculation for the Faraday configuration [9]. For the relative oscillator strengths we use

$$
 f_{\text{rel}} \sim \lim_{r \to 0} \frac{\partial}{\partial r} \left| \left| \langle \pi_x, z | \Psi(r) \rangle \right|^2
$$

for $n = 5$ and with $\varepsilon_{kz} = 6.46$ [20] and $R_{\text{exc}} = 86$ meV [8], i.e., $\hbar K_0$ is the momentum of a photon that has the appropriate energy to create an exciton in the energy range we consider. Since the total mass $M$ of the exciton is some three orders of magnitudes smaller than for a hydrogen atom, this term will have a significant effect on the spectra, even more so if we consider that the region of high fields is shifted to much lower values for the exciton [9]. The term (9) breaks the inversion symmetry and parity ceases to be a good quantum number. While in the Faraday configuration only the dipole-allowed exciton states of odd angular momentum have been important, now also the states of even angular momentum need to be considered. Hence, we need to include the terms for the central cell corrections with the Haken potential as given in Refs. [18, 22] in our treatment to correctly take the coupling to the low lying S states into account.
for light linearly polarized in $x$- or $z$-direction. The states $|\pi_{x,z}\rangle$ are given by

\[
|\pi_x\rangle = \frac{i}{\sqrt{2}} \left[ |2, -1\rangle_{D} + |2, 1\rangle_{D} \right],
\]

\[
|\pi_z\rangle = \frac{i}{\sqrt{2}} \left[ |2, -2\rangle_{D} - |2, 2\rangle_{D} \right],
\]

where $|F_t, M_{F_t}\rangle_{D}$ is an abbreviation [9] for

\[
|(S_e, S_h) S, I; I + S, L; F_t, M_{F_t}\rangle = |(1/2, 1/2) 0, 1; 1, 1; F_t, M_{F_t}\rangle.
\]

In this state, the electron and hole spin $S_e$ and $S_h$ are coupled to the total spin $S$. This is combined first with the quasispin $I$ and then with the orbital angular momentum $L$ to obtain the total angular momentum $F_t$. $M_{F_t}$ is the projection onto the axis of quantization.

**IV. EXPERIMENT**

In the experiment, we investigated the absorption of thin Cu$_2$O crystal slabs. Three different samples with different orientations were available: In the first sample the [001] direction is normal to the crystal surface, in the other two samples the normal direction corresponds to the [110] and [111] orientation, respectively. The thicknesses of these samples differed slightly from 30 to 50 µm which is, however, of no relevance for the results described below. For application of a magnetic field, the samples were inserted at a temperature of 1 K in an optical cryostat with a superconducting split coil magnet. Magnetic fields with strengths up to 7 T could be applied with orientation either parallel to the optical axis (Faraday configuration) or normal to the optical axis (Voigt configuration).

The absorption was measured using a white light source which was filtered by a double monochromator such that only the range of energies in which the exciton states of interest are located was covered. A linear polarization of the exciting light, hitting the crystal normal to the slabs, was used. The transmitted light was dispersed by another double monochromator and detected by a liquid-nitrogen cooled charge coupled device camera, providing a spectral resolution of about 10 µeV. Since the spectral width of the studied exciton resonances is significantly larger than this value, the setup provides sufficient resolution.

**V. RESULTS AND DISCUSSION**

To compare the experimental and numerical spectra in the different configurations we present in Fig. 1 (a) superimposed data with $\sigma^+$- and $\sigma^-$-polarized light in Faraday configuration taken from Ref. [9] and in (b) and (c) spectra in the Voigt configuration with light polarized orthogonally and parallely to the magnetic field axis, respectively, for the principal quantum numbers $n = 4$ and $n = 5$.

Note that we investigate a parameter region where the effects of quantum chaos as discussed in Ref. [26] are not important.

In general, a good agreement between the experimental and numerical data sets is obtained. In the Voigt configuration in panels (b) and (c), a rich splitting is observed, especially of the F-states of the $n = 5$ excitons. We see that light polarized orthogonally to the magnetic field probes complementary lines to the ones excited by light polarized in the direction of the field, a result that will also follow from our discussion below.

In experiment, we are not able to resolve the multiplicity of lines that the calculations reveal. This is related to the increased linewidth of the individual features arising from exciton relaxation by radiative decay and phonon scattering that are not included in the model. Still the field dependences of the main peaks with largest oscillator strength are nicely reproduced as are the broadenings of the multiplets due to level splitting.

**A. Influence of the Magneto-Stark effect**

In this section we want to discuss the effects of the additional effective electric field on the line spectra. As we will see in the following group theoretical derivations, the most pronounced effect is a significant increase in the number of dipole-allowed lines due to the decreased symmetry with the electric field. Panels (a) and (b) in Fig. 1 show this quite clearly, especially for the large number of F-lines. Note that without the Magneto-Stark effect the same selection rules would apply to the spectra in (a) and (b), but not in (c). In contrast to the Faraday configuration [9], we can not limit ourselves to the states with odd values for $L$, owing to the mixture of the even and odd series in the electric field. We discuss the case of a magnetic field aligned in [001] direction and will disregard the influence of the central cell corrections in this discussion.

We consider the reduction of the irreducible representations $D^F_{\pm\pm}$ of the full rotation group in the presence of the crystal as well as the magnetic and effective electric field, where $F = J + L = (I + S_h) + L$ is the angular momentum without the electron spin. With this information we will be able to deduce the splitting of the lines due to the reduced symmetry [27]. Additionally we can compare the resulting irreducible representations with those that the dipole operator belongs to. This will tell us which lines are dipole-allowed and which are not. Note that the symmetry of the quasispin $I$ in $O_h$ is given by $\Gamma_5^+ = \Gamma_4^+ \otimes \Gamma_2^+$ [9] and therefore all irreducible representations have to be multiplied by $\Gamma_2^+$ in comparison with the case of an ordinary spin. Keeping this in mind,
FIG. 1. Comparison of numerical and experimental spectra of the $n = 4$ and $n = 5$ excitons in an external magnetic field $B \parallel [100]$. (a) Superimposed data with $\sigma^+$- and $\sigma^-$-polarized light from Ref. [9] in Faraday configuration. (b) and (c) Voigt configuration with a wavevector aligned with the [001] direction and the light polarized (b) orthogonally and (c) parallelly to the magnetic field. Numerically calculated relative oscillator strengths are shown in grayscale in arbitrary units for a few selected values of $B$ (blue solid lines). To aid comparison, the numerical values are shifted by $-60 \mu$eV whereas the experimental absorption coefficients are lowered with a constant shift to account for phonon background. Numerical spectra were convoluted using a Gaussian function with a constant width of 13.6057 $\mu$eV. Note that the resolution of the numerical data is not uniform for all field strengths. We point out the theoretical visibility of S- and D-excitons as marked for the $n = 4$ lines in (c).

we have [28]

$L = 0$:

$$\tilde{D}^2_{\pm} = D^2_{\pm} \otimes \Gamma_2^+ = \Gamma_0^+ \otimes \Gamma_2^+ = \Gamma_7^+,$$  

(15a)

$L = 1$:

$$\tilde{D}^2_{\pm} = D^2_{\pm} \otimes \Gamma_2^+ = \Gamma_6^+ \otimes \Gamma_2^+ = \Gamma_7^+,$$  

(15b)

$$\tilde{D}^2_{\pm} = D^2_{\pm} \otimes \Gamma_2^+ = \Gamma_6^+ \otimes \Gamma_2^+ = \Gamma_7^+,$$  

(15c)

$L = 2$:

$$\tilde{D}^2_{\pm} = D^2_{\pm} \otimes \Gamma_2^+ = \Gamma_8^+ \otimes \Gamma_2^+ = \Gamma_8^+,$$  

(15d)

$$\tilde{D}^2_{\pm} = D^2_{\pm} \otimes \Gamma_2^+ = (\Gamma_7^+ \otimes \Gamma_8^+) \otimes \Gamma_2^+ = \Gamma_6^+ \otimes \Gamma_8^+,$$  

(15e)

$L = 3$:

$$\tilde{D}^2_{\pm} = D^2_{\pm} \otimes \Gamma_2^+ = (\Gamma_7^+ \otimes \Gamma_8^+) \otimes \Gamma_2^+ = \Gamma_6^+ \otimes \Gamma_8^+,$$  

(15f)

$$\tilde{D}^2_{\pm} = D^2_{\pm} \otimes \Gamma_2^+ = (\Gamma_7^+ \otimes \Gamma_8^+) \otimes \Gamma_2^+ = \Gamma_7^+ \otimes \Gamma_6^+ \otimes \Gamma_8^+,$$  

(15g)

$L = 4$:

$$\tilde{D}^2_{\pm} = D^2_{\pm} \otimes \Gamma_2^+ = (\Gamma_7^+ \otimes \Gamma_8^+) \otimes \Gamma_2^+ = \Gamma_7^+ \otimes \Gamma_6^+ \otimes \Gamma_8^+,$$  

(15h)

$$\tilde{D}^2_{\pm} = D^2_{\pm} \otimes \Gamma_2^+ = (\Gamma_6^+ \otimes \Gamma_8^+) \otimes \Gamma_2^+ = \Gamma_6^+ \otimes \Gamma_6^+ \otimes \Gamma_8^+,$$  

(15i)

We still need to include the spin of the electron which transforms according to $\Gamma_6^\uparrow$. For vanishing magnetic field strengths, the representations belonging to an irreducible representation without the spin are degenerate. Those will be written in brackets. The reduction [28] will only be specified for even parity, since the odd case only changes the sign. We obtain

$$\tilde{D}^2_{\pm} \otimes \Gamma_6^+ = (\Gamma_2^+ \otimes \Gamma_6^+),$$  

(16a)

$$\tilde{D}^2_{\pm} \otimes \Gamma_6^+ = (\Gamma_3^+ \otimes \Gamma_4^+ \otimes \Gamma_6^+),$$  

(16b)

$$\tilde{D}^2_{\pm} \otimes \Gamma_6^+ = (\Gamma_1^+ \otimes \Gamma_4^+ \otimes \Gamma_6^+) \otimes (\Gamma_3^+ \otimes \Gamma_4^+ \otimes \Gamma_5^+),$$  

(16c)

$$\tilde{D}^2_{\pm} \otimes \Gamma_6^+ = (\Gamma_1^+ \otimes \Gamma_5^+) \otimes (\Gamma_1^+ \otimes \Gamma_4^+) \otimes (\Gamma_3^+ \otimes \Gamma_4^+ \otimes \Gamma_5^+),$$  

(16d)
\[ \hat{D}^2 \gamma \otimes \Gamma_6^+ = (\Gamma_3^+ \oplus \Gamma_5^+) \oplus (\Gamma_4^+ \oplus \Gamma_1^-) \]
\[ \oplus (\Gamma_7^+ \oplus \Gamma_4^- \oplus \Gamma_5^-) \]  

(16c)

\[ \Gamma_1^+ \text{ and } \Gamma_2^+ \text{ are one-dimensional, } \Gamma_3^+ \text{ is two-dimensional, and } \Gamma_4^+ \text{ and } \Gamma_5^+ \text{ are three-dimensional. So without the field, we have for example fourfold degenerate S-states and P-states that are split into one fourfold and one eightfold degenerate line. If the magnetic field is switched on, the electric field becomes nonvanishing too. The symmetry is reduced from } O_h \text{ to } C_S \text{ [28]. All representations of } C_S \text{ are one-dimensional, so all degeneracies will be lifted, just as in the case with only a magnetic field. But in contrast to the Faraday configuration, the symmetry is lowered even further, leading to a greater mixture of the states. In fact, all lines become dipole-allowed. To see this, we have to consider the reduction of the irreducible representations of } O_h \text{ in } C_S \text{ [27, 28]. The relevant expressions are}

\[
\begin{align*}
\Gamma_1^+ & \to \Gamma_1, & \Gamma_3^- & \to \Gamma_2, \\
\Gamma_2^+ & \to \Gamma_1, & \Gamma_2^- & \to \Gamma_2, \\
\Gamma_3^+ & \to \Gamma_1 \oplus \Gamma_1, & \Gamma_3^- & \to \Gamma_2 \oplus \Gamma_2, \\
\Gamma_4^+ & \to \Gamma_1 \oplus \Gamma_2 \oplus \Gamma_2, & \Gamma_4^- & \to \Gamma_2 \oplus \Gamma_1 \oplus \Gamma_1, \\
\Gamma_5^+ & \to \Gamma_1 \oplus \Gamma_2 \oplus \Gamma_2, & \Gamma_5^- & \to \Gamma_2 \oplus \Gamma_1 \oplus \Gamma_1.
\end{align*}
\]

The dipole operator belongs to } \Gamma_1^- \text{ in } O_h \text{ [28] and its reduction therefore includes all appearing representations. Thus, all } 4n^2 \text{ lines receive nonvanishing oscillator strength, the only limitation being given by the polarization of the incident light, i.e., a given state can either be excited by radiation polarized in the } z\text{-direction (} \Gamma_2) \text{ or by radiation polarized in the } x\text{-y-plane (} \Gamma_1) \text{.}

\section{VI. SUMMARY}

We extended the previous work by Schweiner et al. [9] on the optical spectra of magnetoexcitons in cuprous oxide to the Voigt configuration and showed that the nonvanishing exciton momentum perpendicular to the magnetic field leads to the appearance of an effective Magneto-Stark field. Including the valence band structure and taking into account central cell corrections as well as the Haken potential allowed us to produce numerical results in good agreement with experimental absorption spectra. We observe a significant increase in the number of visible lines in both our experimental as well as our numerical data as compared to the Faraday configuration. Using group theoretical methods, we show that this is related to the Magneto-Stark field increasing the mixing between states. While their positions remain relatively unaffected, the mixing of states leads to finite oscillator strength of, at least in principle, all lines.

\section{ACKNOWLEDGMENTS}

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