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# Anisotropic effective exciton mass in $Cu_2O$

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The degeneracy of the yellow 1S orthoexcitons in $Cu_2O$ is lifted by wave vector dependent electron-hole exchange. This interaction scales quadratically with $k$ and strongly depends on the direction of the wave vector. Therefore it is interpreted as an anisotropic correction of the effective exciton mass. This can explain the discrepancies in the measurements of the exciton mass in $Cu_2O$ reported so far.



Spins are coupled via exchange interaction. For exciton physics the exchange between the coupled electron-hole pairs is of increased interest. Despite being studied intensively the understanding of electron-hole exchange even in bulk materials is far from being complete. The electron-hole exchange interaction depends on the full band structure and hence on the wave vector $\mathbf{k}$ of the exciton center-of-mass motion. However, normally it is approximated as a spin-spin interaction, which is independent of the wave vector. This represents a severe simplification of the exchange interaction.

Recently the wave-vector-dependence of the electron-hole exchange has been derived and experimentally verified for the yellow 1s orthoexciton in $Cu_2O$.[1] In this article we will summarize the essential results and discuss the impact of wave-vector-dependent exchange for the interpretation of the effective exciton mass.

In $Cu_2O$ excitonic transitions with holes in the $\Gamma_7^+$ band and electrons in the $\Gamma_6^+$ band give the so-called yellow exciton series. One finds four 1S excitons that are split by $k$-independent exchange into the threefold $\Gamma_5^+$ states termed orthoexcitons and a single $\Gamma_2^+$ level referred to as paraexciton. In the following we will solely concentrate on the orthoexcitons. In lowest order, the orthoexcitons couple to light via quadrupole interaction. Exchange interaction of order $k^0$ leaves the orthoexcitons degenerate. When calculating exchange of higher order in $k$ one finds that the $k$-linear terms vanish. The calculation of the terms of order $k^2$ is described in great detail in Ref. [1, 2]. Summarizing one finds the long range exchange term

$$J_{ex}^{Q} = \Delta_Q \cdot \frac{1}{k^2} \begin{pmatrix} k_z^2 k_y^2 & k_z^2 k_x k_y & k_y^2 k_z k_x \\ k_z^2 k_x k_y & k_z^2 k_x^2 & k_x^2 k_y k_z \\ k_y^2 k_z k_x & k_x^2 k_y k_z & k_x^2 k_y^2 \end{pmatrix}. \tag{1}$$

In contrast to dipole allowed excitons, the long range part of the quadrupole-quadrupole exchange scales as $k^2$ and is analytic at $k = 0$.

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Further one obtains three short range terms:

$$J_1 = \Delta_1 \cdot \begin{pmatrix} k^2 & 0 & 0 \\ 0 & k^2 & 0 \\ 0 & 0 & k^2 \end{pmatrix} = \Delta_1 k^2 \cdot \mathbf{1}, \tag{2}$$

$$J_3 = \Delta_3 \cdot \begin{pmatrix} 3k_x^2 - k^2 & 0 & 0 \\ 0 & 3k_y^2 - k^2 & 0 \\ 0 & 0 & 3k_z^2 - k^2 \end{pmatrix}, \tag{3}$$

$$J_5 = \Delta_5 \cdot \begin{pmatrix} 0 & k_x k_y & k_x k_z \\ k_x k_y & 0 & k_y k_z \\ k_x k_z & k_y k_z & 0 \end{pmatrix}. \tag{4}$$

$J_1$ is proportional to the identity matrix and thus causes an isotropic spectral shifts for all orthoexcitons. The $J_3$ contribution is also diagonal. However, the different $\mathbf{k}$-dependencies of the diagonal elements give rise to an orthoexciton fine structure. The off-diagonal $J_5$ mixes the $\Gamma_5^+$ states. The exchange parameters $\Delta_Q$, $\Delta_3$, and $\Delta_5$ were determined in high resolution transmission experiments [1, 2]. The $k$-dependence is evaluated by investigating crystals of various orientations. Using this technique, one finds $\Delta_Q$ = 5 $\mu$eV, $\Delta_3$ = -1.3 $\mu$eV and $\Delta_5$ = 2 $\mu$eV. The exchange parameter $\Delta_1$ can not be determined directly, as it does not give rise to a splitting among the orthoexciton states. The resulting fine structure for $\mathbf{k}$'s in the [001]-[110] plane is shown in Fig. 1 (a). The solid symbols mark experimental values. The energy shifts $\Delta E$ include the exchange contributions $J_{ex}^Q$, $J_3$, and $J_5$

One might argue that the exchange contributions of order $k^2$ are relatively small. They are about three orders of magnitude smaller than the conventional $k$-independent exchange between ortho-and paraexcitons. However these small contributions can be of crucial importance for example for the effective mass of the exciton $M$.

The exchange terms discussed here scale quadratically in $k$ and can therefore be interpreted in terms of a direction dependent, anisotropic effective mass. Naturally, previous measurement of $M$ have not taken these contributions into account. As we have seen, the exchange shifts are on the order of few $\mu$eV and hence comparable to the kinetic energy of the exciton at the exciton-photon resonance $|\mathbf{k}| = k_0 = 2.62 \cdot 10^7$ m$^{-1}$.

As pointed out already by Yu and Shen [3, 4], there is a large discrepancy between the effective exciton mass $M_0$ derived from the sum of the band masses of the conduction band $m_e$ and the valence band $m_h$ ($M_0 = m_e + m_h$) and the exciton mass derived from resonant Raman experiments. From the latter the authors obtain $M = (3.0 \pm 0.2)\, m_0$, with the free electron mass $m_0$. From cyclotron resonance experiments Goltzene and Schwab find the band masses $m_e = 0.98\ m_0$ and $m_h = 0.66\ m_0$ [5]. Having in mind the wave vector dependent exchange the exciton mass has to be interpreted as follows: We assume the crystal in the resonant Raman experiments were oriented along $\mathbf{k} = [001]$. For this $\mathbf{k}$ only the degenerate $E_{1,3}$ level is optically allowed, which means that the investigated orthoexciton is shifted by $-\Delta_3$ from our reference energy $E_0$. This reference energy includes all isotropic shifts of the exciton. Hence it already includes the exchange term $J_1$ with the so far unknown exchange energy $\Delta_1$ and the kinetic energy of the electron and the hole.

In consequence, for the measured effective exciton mass for $\mathbf{k} = [001]$ ($M_{1,3} = 3.0\ m_0$), the $J_1$ and $J_3$ terms have to be included. The effective mass $M_{1,3}$ of the $E_{1,3}$ resonance is then given by

$$\frac{\hbar^2 k_0^2}{2M_{1,3}} = \frac{\hbar^2 k_0^2}{2M_0} + \Delta_1 - \Delta_3. \tag{5}$$

As we also know the individual masses of the exciton constituents and $\Delta_3$, we can now determine $\Delta_1 = -8.6$ $\mu$eV. The exchange interaction between the exciton constituents gives thus rise to an effective mass significantly different from the sum of the electron and hole mass. Taking the exchange into account,

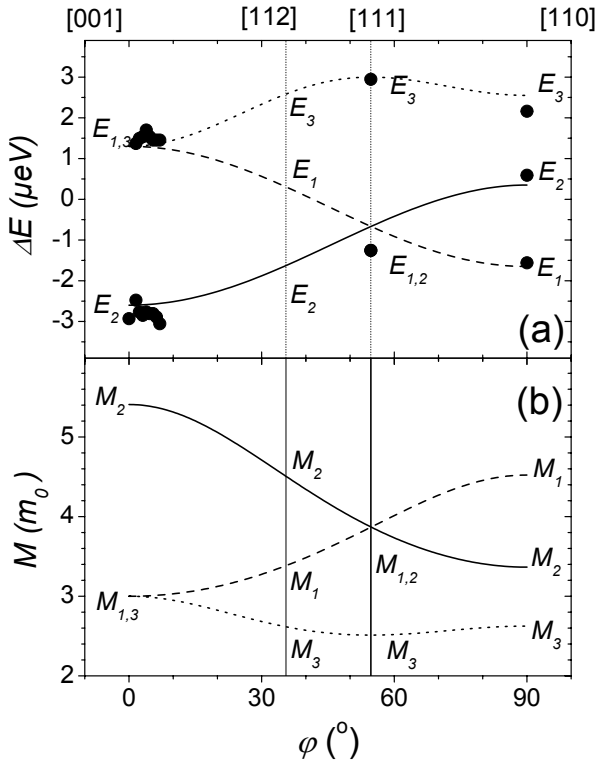


Fig. 1: (a) Exchange shifts of the orthoexcitons as function of $\varphi$. $\mathbf{k}$ lies in the $[001]$-$[110]$ plane. Solid symbols indicate measured exchange splittings. (b) Effective exciton mass as function of $\varphi$.

the controversial reports on the exciton mass can be explained in a straight forward way. However, also central-cell corrections might be another reason for this difference [6].

Obviously the $\mathbf{k}$-dependence of the exchange exchange terms $J_{ex}^{Q}$, $J_3$, and $J_5$ gives rise to an anisotropy of the effective mass. Using Eq. (5) and knowing the anisotropic exchange shifts $\Delta E_i(\mathbf{k})$ we can now calculate the effective mass $M_i$ of each orthoexciton $i$ as function of $\mathbf{k}$:

$$\frac{1}{M_i(\mathbf{k})} = \frac{1}{M_{1,3}(\mathbf{k}=[001])} + \frac{2}{\hbar^2 k_0^2}(\Delta_3 + \Delta E_i(\mathbf{k})). \tag{6}$$

Fig. 1 (b) shows the exciton masses for the $\mathbf{k}$-directions in the $[001]-[110]$ plane. Indeed the anisotropy is significant. For example at $\mathbf{k}=[001]$ the effective mass of the two orthoexcitons differs by almost a factor of two.

In Fig. 2 the anisotropy of the dispersion is illustrated for $\mathbf{k}_{110}$ (panel (a)) and $\mathbf{k}_{001}$ (panel (b)). For comparison also the dispersion, as expected for the bare band masses of the exciton components is shown. In the present experiment the exchange splittings can only be measured at the intersections of the exciton dispersions with the light cone. The exchange parameters $\Delta_i$ are measured for $k_0$ and are expected to scale as $\Delta_i k^2/k_0^2$. The orthoexcitons are again degenerate at $k=0$. This could be verified in a high resolution two photon experiment with counter propagating laser beams.

Another aspect should be mentioned here. The yellow 1S ortheoexcitons in $Cu_2O$ play a key role in many proposals and experimental investigations of Bose-Einstein-condensation of excitons. The critical density for such condensation phenomena is a crucial parameter. This density scales linearly with the degeneracy of the exciton state. In the absence of perturbations like strain or magnetic fields one has treated up to now the orthoexcitons as threefold degenerate. However, when taking the $k^2$-exchange into

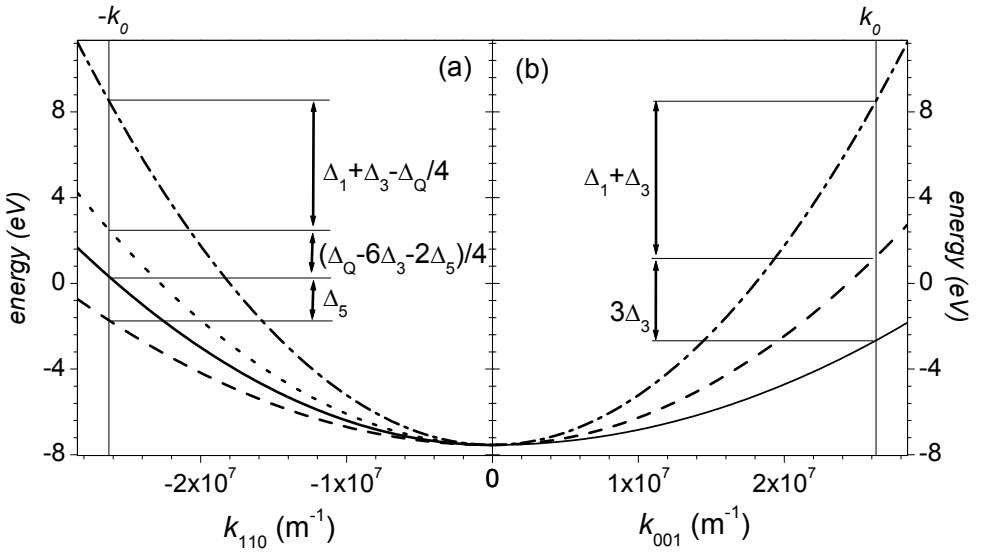


Fig. 2: Orthoexciton fine structure as function of $k$ for $\mathbf{k}_{110}$ (a) and $\mathbf{k}_{001}$ (b). The dash dotted trace gives the dispersion as expected from the sum of the band masses $m_e$ and $m_h$. $k_0$ marks the intersection with the light cone. (a) The dashed (dotted) line gives $E_1$ ($E_3$) and the solid line gives $E_2$.(b) The dashed (solid) line gives $E_{1,3}$ ($E_2$) (compare Fig. 1 (a)).

account, the degeneracy of the orthoexcitons can be lifted for $k \neq 0$. Hence the critical density would be by a factor of $\leq 3$ smaller than previously assumed.

Even though the orthoexcitons couple to photons only via quadrupole interaction, this still gives rise to pronounced polaritonic behavior [7]. Calculations show that this prevents a steady state Bose-Einstein condensation of orthoexcitons [8]. However, using the exchange splitting and the polarization dependence of the quadrupole transition one can selectively study an exciton component with reduced oscillator strength. This allows to reduce the polaritonic coupling, which will be advantageous in the search for exciton condensation.

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