Interseries dipole transitions from yellow to green excitons in cuprous oxide

Patric Rommel and Jörg Main
Institut für Theoretische Physik 1, Universität Stuttgart, 70550 Stuttgart, Germany

Sjard Ole Krüger and Stefan Scheel
Institut für Physik, Universität Rostock, Albert-Einstein-Straße 23-24, 18059 Rostock, Germany
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We study dipole interseries transitions between the yellow and green exciton series in cuprous oxide including the complex valence band structure. To this end, we extend previous studies of the spectrum of complex green exciton resonances [Phys. Rev. B 101, 075208 (2020)] to optical transitions between different exciton states in addition to transitions from the crystal ground state. This allows us to augment the calculations on interseries transitions using a hydrogen-like model [Phys. Rev. B 100, 085201 (2019)] by a more comprehensive treatment of the valence band structure.

I. INTRODUCTION

Cuprous oxide has long been a very interesting system for the study of excitons. Indeed, it was in this material that excitons were first observed [1, 2], and in which bound states with principal quantum numbers up to \( n = 25 \) have been detected [3]. This abundance of known resonances allows for very precise tests of theoretical models, and to probe the influence of intricate details of the band structure on the formation of excitons.

Most of the work in the literature focuses on the yellow series, which is formed by electrons in the lowest \( \Gamma_6^+ \) conduction band and holes in the uppermost \( \Gamma_7^+ \) valence band [4–6]. The green excitons, on the other hand, are formed by holes in the \( \Gamma_8^+ \) valence band [7–9]. For principal quantum numbers \( n \geq 2 \), they are located at energies above the band gap of the yellow excitons, and couple to the yellow continuum states. Thus, even without taking phonon coupling into account, the green excitons above the yellow band gap are no longer bound states with infinite lifetimes, but quasi-bound resonances with finite lifetimes. Recently, the locations of the green exciton resonances have been calculated [10] using the complex-scaling method [11, 12].

Motivated by the aim to identify promising experimentally accessible dipole transitions for the coherent manipulation of Rydberg excitons [13–15] and the generation of giant optical nonlinearities [15], interseries transitions between the yellow and green, respectively yellow and blue, exciton series have been investigated using a hydrogen-like model for the exciton interaction [16]. The interseries transitions, i.e., those between different exciton series, have the distinct advantage over intraseries transitions, i.e., transitions within a single exciton series, by providing a more accessible choice of interrogation wavelengths. To wit, transition wavelengths between adjacent Rydberg states within the same series scale as \( n^3 \) and quickly approach the millimeter range, whereas the wavelength limit for interseries transitions is set by the energy difference between bands, which are typically in the near- to mid IR. In this article, we investigate interseries dipole transitions between the yellow and green exciton series, while taking into account the complex structure of the valence band [5, 17] as well as central-cell corrections [18–22]. As our focus is not on optical transitions where the exciton is created from the crystal ground state, but rather on transitions between different exciton states, this requires an extensive modifications of the scheme for calculating the oscillator strengths.

The article is organized as follows. First, we present the numerical calculation of the relevant exciton states using a unified Hamiltonian describing both the yellow and green series in Sec. II. Using the calculated eigenvalues and eigenvectors, we derive the dipole transition matrix elements in Sec. III. In Sec. IV we present and discuss our results on interseries dipole transitions and absorption spectra. We finish the paper with some concluding remarks in Sec. V.

II. THE SPECTRUM OF YELLOW AND GREEN EXCITONS

In this section we briefly recapitulate and extend our technique for calculating the bound yellow exciton states and the unbound green exciton resonances by using a complete basis set and the complex-coordinate rotation method. For both the yellow and green series, we take the valence band structure and central-cell corrections into account. The knowledge of the precise states is the prerequisite for the computation of interseries dipole transitions in Secs. III and IV.

A. Hamiltonian

The description of excitons follows the line of arguments laid out in Ref. [10]. For the investigation of interseries transitions between yellow and green exciton states, we use the unified description of the two series.
given by the Hamiltonian \[ H = E_g + \frac{\gamma_1}{2m_0} p^2 + H_b(p) - \frac{e^2}{4\pi\varepsilon_0\varepsilon} |r| + H_{CCC}, \] with the valence-band corrections to the kinetic energy \[ H_b(p) = H_{SO} + \frac{1}{2\hbar^2m_0} \left\{ 4\hbar^2\gamma_2 p^2 + 2(\eta_1 + 2\eta_2)p^2(I \cdot S_h) - 6\gamma_2(p_1^2 I_1^2 + \text{c.p.}) - 12\eta_2(p_1^2 I_1 S_{h1} + \text{c.p.}) - 12\gamma_3((p_1, p_2)\{I_1, I_2\} + \text{c.p.}) - 12\eta_3((p_1, p_2)\{I_1 S_{h2} + I_2 S_{h1}\} + \text{c.p.}) \right\}, \] and the central-cell corrections \( H_{CCC} \) discussed below in Sec. II C. Here we use center-of-mass coordinates, \( r, p = \frac{hr_e + m_e r_h}{m_h + m_e} \), \( P = \hbar K = p_e + p_h \), \( \rho = \hbar k = \frac{m_h p_e - m_e p_h}{m_h + m_e} \), \[ \gamma_i = \gamma_1 + m_0/m_e, \quad E_g \text{ is the gap energy, } \varepsilon \text{ the dielectric constant, and } c.p. \text{ denotes cyclic permutation.} \]

The band structure Hamiltonian \( H \) reduces complex eigenenergies, whose imaginary part is related to the phonons. For their description, we introduce complex-valued eigenenergies, as schematically illustrated in Fig. 1. Continuum states are rotated into the lower complex energy plane, revealing the resonances, which are hidden in a Hermitian eigenvalue problem. If the rotation angle \( \vartheta \) is chosen appropriately, the resonance states become square integrable. For additional details we refer the reader to Refs. [10, 12].

C. Central-cell corrections

For a correct description of the even-parity exciton states, additional central-cell corrections \[ \Delta = -2 \text{Im} E. \] under the complex-coordinate rotation, the Hamiltonian \( H \) becomes a non-Hermitian operator, and thus allows for complex-valued eigenenergies, as schematically illustrated in Fig. 1. Continuum states are rotated into the lower complex energy plane, revealing the resonances, which are hidden in a Hermitian eigenvalue problem. If the rotation angle \( \vartheta \) is chosen appropriately, the resonance states become square integrable. For additional details we refer the reader to Refs. [10, 12].

B. Complex-coordinate rotation

The green exciton states lie above the band gap of the yellow series and are coupled to the yellow continuum. Hence, they form quasi-bound resonances rather than bound states, even without considering the coupling to the phonons. For their description, we introduce complex eigenenergies, whose imaginary part is related to the finite linewidth as \( \gamma = -2 \text{Im} E. \) To compute these eigenenergies, we perform the complex-coordinate rotation \( r \rightarrow re^{i\vartheta} \). It is important to note that, under the complex-coordinate rotation, the Hamiltonian \( H \) becomes a non-Hermitian operator, and thus allows for complex-valued eigenenergies, as schematically illustrated in Fig. 1. Continuum states are rotated into the lower complex energy plane, revealing the resonances, which are hidden in a Hermitian eigenvalue problem. If the rotation angle \( \vartheta \) is chosen appropriately, the resonance states become square integrable. For additional details we refer the reader to Refs. [10, 12].

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in the Hamiltonian $H$ are necessary. Here, the Haken potential

$$V^\text{HI} = -\frac{e^2}{4\pi\epsilon_0 r} \left[ \frac{1}{2\epsilon_1} \left( e^{-\frac{r}{2\epsilon_1}} + e^{-\frac{r}{\epsilon_1}} \right) + \frac{1}{2\epsilon_2} \left( e^{-\frac{r}{2\epsilon_2}} + e^{-\frac{r}{\epsilon_2}} \right) \right]$$

(6)

describes corrections to the dielectric constant for small exciton radii,

$$V_d = -V_0 V_{uc} \delta(r)$$

(7)
is an additional short distance correction \cite{22}, and

$$H_{\text{exch}} = J_0 \left( \frac{1}{4} - \frac{1}{\hbar^2} S_e \cdot S_h \right) V_{uc} \delta(r)$$

(8)
is the exchange interaction \cite{20}, which causes a splitting of the $S$-type states into ortho- and paraexcitons depending on the relative orientation of the electron and hole spins. For the central-cell corrections, we introduce the polaron radii

$$\rho_{e/h,i} = \sqrt{\frac{\hbar}{2m_e / \hbar^2 \omega_{\text{LO},i}}}$$

(9)

with the energies $\hbar \omega_{\text{LO},i}$ of the longitudinal $\Gamma_4$ phonons, and the values

$$\frac{1}{\epsilon_1^2} = \frac{1}{\epsilon_{bi}} - \frac{1}{\epsilon_{si}}.$$ 

(10)

The parameters $J_0$ and $V_0$ are given in Table 1. $V_{uc} = a^3$ is the volume of the unit cell.

Due to the cubic crystal symmetry and the associated coupling to angular-momentum states with $\Delta l = \pm 2$, these corrections affect not only the $S$ states but also the other even-parity states. The implementation of these terms requires the calculation of the complex-rotated matrix elements given in Appendix D of Ref. \cite{18}. The difficulty is that these matrix elements form an alternating sum of terms with individually very large absolute values. Thus, an accurate calculation requires the use of a large number of significant digits. We therefore work with a computer algebra system to perform computations to arbitrary precision instead of standard double-precision calculations. With these preliminaries, we are now in the position to calculate the spectrum of even-parity green exciton states.

D. Non-Hermitian generalized eigenvalue problem

To calculate the eigenstates and eigenvalues of the Hamiltonian $H$, we express the time-independent Schrödinger equation in a complete basis $|\Pi\rangle$ using basis states

$$|\Pi\rangle = |N, L, (I, S_h), J, F, S_e, F_1, M_{F_1}\rangle$$

(11)

with orbital angular momentum $L$, effective hole spin $J$ (as the sum of the quasispin $I$ and the hole spin $S_h$), angular momentum $F = J + L$, and total angular momentum $F_1 = F + S_e$ with its $z$-component $M_{F_1}$. Here, $S_e$ denotes the electron spin. For the radial part, we use complex rotated Coulomb-Sturmian functions \cite{31},

$$U_{NL}(r) = N_{NL}(2r/\alpha)^{\frac{3}{2}} e^{-r/\alpha} L_N^{2l+1}(2r/\alpha),$$

(12)

which depend on $L$, and are additionally characterized by the radial quantum number $N$ and the convergence parameter $\alpha$. The latter can be used for the implementation of the complex scaling operation, allowing for the calculation of complex resonance states. To this end, a complex-valued $\alpha = |\alpha| e^{i\theta}$ is chosen, resulting in the complex rotation with angle $\theta$.

When expressing the exciton states $|\Psi\rangle$ in the basis $|\Pi\rangle$,

$$|\Psi\rangle = \sum_{\Pi} c_{\Pi} |\Pi\rangle,$$

(13)

the Schrödinger equation becomes a non-Hermitian generalized eigenvalue problem,

$$Ac = EMc$$

(14)

with the Hamiltonian matrix $A_{\Pi\Pi} = \langle \Pi | H | \Pi \rangle$, the overlap matrix $M_{\Pi\Pi} = \langle \Pi | | \Pi \rangle$ and the vector $c$ containing the coefficients $c_{\Pi}$. Note that the overlap matrix $M$ differs from the identity because the Coulomb-Sturmian functions $|\Pi\rangle$ are not orthogonal. To obtain finite matrices and vectors, we introduce cut-offs to the quantum numbers $N + L + 1 \leq n_{\text{max}}$ and $F \leq F_{\text{max}}$. These parameters, together with $|\alpha|$ and $\theta$ have to be chosen appropriately to ensure properly converged results. Good convergence is reached when variations of the parameters do not lead to significant changes in the calculated spectra.

We first diagonalize the Hamiltonian excluding the singular Dirac delta-terms $V_d$ and $H_{\text{exch}}$ of the central-cell corrections \cite{5}. From the high-dimensional matrices, we are only interested in a small window of eigenstates. For this aim an iterative method is implemented (e.g. in the ARPACK package \cite{72}) that allows for the calculation of eigenvalues and eigenvectors near a controllable predetermined energy, which is numerically more efficient than a direct diagonalization.

After this, we set up a second eigenvalue problem where we include the delta-terms with only the converged eigenstates from the first diagonalization. For this, we diagonalize the entire resulting low-dimensional eigenvalue problem using a direct LAPACK method \cite{33}.

III. DIPOLE TRANSITIONS BETWEEN EXCITONIC STATES

In the following, we investigate dipole transitions between different exciton state which is in contrast to earlier
work that focused mostly on transitions from the crystal ground state [5,11,18]. The central quantity describing the transition from an initial exciton state $|\Psi_i\rangle$ to the final exciton state $|\Psi_f\rangle$ is the transition matrix element

$$M_{\text{fi}} = \langle \Psi_f | \hat{e}_A \cdot \mathbf{\pi} | \Psi_i \rangle$$  \hspace{1cm} (15)$$

of the single-photon transition operator, with the polarization direction $\hat{e}_A$ of the vector potential associated with the photon field

$$A(x) = A_0 e^{i k_0 x} = A_0 = A_0 \hat{e}_A$$  \hspace{1cm} (16)$$
in dipole approximation, where we assume that the momentum $h k_0$ of the photon is much smaller than the relative momentum of exciton and hole. The operator

$$\mathbf{\pi} = m_0 \mathbf{v} = m_0 \frac{\partial \mathbf{x}}{\partial t} = \frac{im_0}{\hbar} [\mathcal{H}, \mathbf{x}]$$  \hspace{1cm} (17)$$
denotes the kinetic momentum operator in a crystal with spin-orbit interaction, and appears during the minimal-substitution procedure. Note that it differs from the quasi-momentum $p$ associated with the Bloch eigenfunctions of the band Hamiltonian $\mathcal{H}$. The position operator $\mathbf{x}$ also has to be distinguished from the coordinates $r_e$ and $r_h$ that arise from the lattice positions in the continuum description of the crystal.

For the interseries transitions discussed in this paper, $|\Psi_i\rangle$ is mostly a bound yellow exciton state and $|\Psi_f\rangle$ is an unbound green exciton resonance. It is therefore sufficient to consider the matrix elements in a basis, e.g., with the basis states $|\Pi\rangle$; the transition amplitudes for the eigenstates can then be obtained by forming appropriate superpositions,

$$M_{\text{fi}} = \sum_{\Pi':\Pi} \langle \Pi' | \hat{e}_A \cdot \mathbf{\pi} | \Pi \rangle.$$  \hspace{1cm} (18)$$

Note that the coefficients for the left (bra vector) basis states are not complex conjugated, since they would be real valued without the complex-coordinate rotation.

A. Operator identity between kinetic momentum and derivatives of the band Hamiltonian

We will now derive an operator identity between the kinetic momentum $\mathbf{\pi}$ and the derivatives of the band Hamiltonian $H(p)$ with respect to the momenta. For that, we consider arbitrary single exciton states, which are effective two-particle states of an electron with spin $S_{e,z} = \sigma_e$ in the conduction band (c) and a hole with effective hole spin $J_{h,z} = \sigma_h$ in the valence band (v). An excitonic state with center-of-mass momentum $P$ can then be written as

$$|\Psi_{c,v}^{\tau,p}\rangle = \sum_p \phi_{\tau,p}(p) a_{c,\sigma_e,p+\alpha_h,p} b_{v,\sigma_h,-p+\alpha_h,p} |\Psi_0\rangle$$  \hspace{1cm} (19)$$

where $\tau = \{N,L,M,\sigma_e,\sigma_h\}$ is a shorthand notation for all the additional quantum numbers of the exciton, $a_{c,\sigma_e,p}^\dagger$ and $b_{v,\sigma_h,p}^\dagger$ denote an electron (hole) creation operator and $|\Psi_0\rangle$ the crystal ground state. The coefficients $\alpha_e$ and $\alpha_h$ stem from the transformation from electron-hole coordinates to relative and center-of-mass coordinates. They are in principle arbitrary but must fulfill $\alpha_e + \alpha_h = 1$. We have chosen the same coefficients for all states.

1. Dipole approximation

In the dipole approximation, the excitonic center-of-mass momentum $P$ vanishes and Eq. (19) reduces to

$$|\Psi_{c,v}^{\tau}\rangle = \sum_p \phi_{\tau,p}(p) a_{c,\sigma_e,p}^\dagger b_{v,\sigma_h,-p} |\Psi_0\rangle.$$  \hspace{1cm} (20)$$
The single-photon transition operator in dipole approximation, projected onto the Hilbert space spanned by the states in Eq. (20), can be written as

$$\frac{e A_0 \pi}{m_0} = \frac{e A_0}{m_0} \sum_{\nu,\nu'} \sum_{\sigma_e,\sigma_v} \sum_q \langle \nu, \sigma_e, q | \pi | \nu', \sigma_v, q \rangle a_{\nu,\sigma_e,q}^\dagger a_{\nu',\sigma_v,q} + \frac{e A_0}{m_0} \sum_{\xi,\xi'} \sum_{\sigma_h,\sigma_v} \sum_q \langle \xi, \sigma_h, q | \pi | \xi', \sigma_v, q \rangle b_{\xi,\sigma_h,q}^\dagger b_{\xi',\sigma_v,q}$$  \hspace{1cm} (21)$$

where $A_0$ denotes the vector potential [16], the indices $\nu, \nu'$ sum over all conduction bands, the indices $\xi, \xi'$ sum over all valence bands and the $\sigma_{e/h}$ denote the corresponding substates (spins). In the derivation of Eq. (21), the Coulomb gauge has been used and the diamagnetic term was ignored, as it only has an appreciable influence for very high field strengths of the incoming electromagnetic wave. Upon evaluating a matrix element of the kind $e A_0 \langle \Psi_{c,v}^{\tau}; | \pi | \Psi_{c,v}^{\tau'}\rangle / m_0$, there are four cases that must be analyzed separately.

(i) Intraseries transitions: Here, we have $\{c,\sigma_e\} = \{c',\sigma'_{e}\}$ and $\{v,\sigma_h\} = \{v',\sigma'_{h}\}$. Applying the fermionic anti-commutation rules for the creation and annihilation operators, one arrives at

$$\langle \Psi_{c,v}^{\tau'} | \pi | \Psi_{c,v}^{\tau}\rangle = \sum_p \phi_{\tau',p}(p) \phi_{\tau,p}^\dagger(p)$$  \hspace{1cm} (22)$$

$$\times (\langle c, \sigma_e, p | c, \sigma_e, p \rangle | v, \sigma_h, -p \rangle | v, \sigma_h, -p \rangle - \langle v, \sigma_h, -p | v, \sigma_h, -p \rangle).$$

(ii) Hole-driven interseries transitions: In this case, we have $\{c,\sigma_e\} = \{c',\sigma'_{e}\}$ but $\{v,\sigma_h\} \neq \{v',\sigma'_{h}\}$. Hence, we arrive at

$$\langle \Psi_{c,v}^{\tau'} | \pi | \Psi_{c,v}^{\tau'}\rangle = \sum_p \phi_{\tau',p}(p) \phi_{\tau,p}^\dagger(p) \langle v, \sigma_h, -p | \pi | v', \sigma'_h, -p \rangle.$$  \hspace{1cm} (23)$$
(iii) Electron-driven interseries transitions: Here, we have \( \{ c, \sigma_e \} \neq \{ c', \sigma'_e \} \) but \( \{ v, \sigma_h \} = \{ v', \sigma'_h \} \). This case yields

\[
\langle \Psi_{c',v} | \Psi_{c,v} \rangle = \sum_p \phi_{c'}(p) \phi_{c'}^*(p) \langle c, \sigma_e, p | c', \sigma'_e, p \rangle.
\]

(24)

(iv) Two-particle transitions: In this case, one has \( \{ c, \sigma_e \} \neq \{ c', \sigma'_e \} \) and \( \{ v, \sigma_h \} \neq \{ v', \sigma'_h \} \). These transitions are forbidden to all orders in single-photon transitions and will not be discussed further.

The transitions from the yellow to the green series in Cu2O are predominantly hole-driven, although there might be admixture of yellow states into the green series and vice versa.

2. Bloch matrix elements

The interband matrix elements \( \langle n, \sigma, p | \pi | n', \sigma', p \rangle \), which are expressed in terms of Bloch states, can be written in terms of the lattice periodic functions \( | u_{n,\sigma}, p \rangle \) via

\[
\langle n, \sigma, p | \pi | n', \sigma', p \rangle = e^{i\mathbf{kr}} | u_{n',\sigma}, p \rangle \text{ which results in}
\]

\[
\langle n, \sigma, p | \pi | n', \sigma', p \rangle = \langle u_{n,\sigma}, p | \pi | u_{n',\sigma'}, p \rangle + \mathbf{p} \delta_{n,n'} \delta_{\sigma,\sigma'}.
\]

(25)

Here \( n, n' \) denote the bands and \( \sigma, \sigma' \) the associated spins. The Hamiltonian acting on these lattice periodic functions is the \( \mathbf{p} \cdot \pi \)-Hamiltonian (usually referred to as the \( \mathbf{k} \cdot \pi \)-Hamiltonian with \( \mathbf{k} = p/\hbar \))

\[
\mathcal{H}_{\mathbf{p} \cdot \pi} = \mathcal{H}_0 + \mathcal{H}_p,
\]

(26)

with

\[
\mathcal{H}_0 = -\frac{\hbar^2 \nabla^2}{2m_0} + V(x) - \frac{i\hbar^2}{4m_0^2 c^2} (\mathbf{\sigma} \times \nabla V(x)) \cdot \nabla,
\]

(27)

\[
\mathcal{H}_p = \frac{\mathbf{p}}{m_0} \cdot \pi + \frac{\mathbf{p}^2}{2m_0},
\]

(28)

where \( V(x) \) is the lattice periodic potential, \( \mathbf{\sigma} \) the vector of Pauli matrices, and \( \mathcal{H}_0 \) denotes the Hamiltonian at the \( \Gamma \)-point. This implies the relation

\[
\pi = m_0 \mathbf{v} = m_0 \frac{\partial \mathcal{H}_{\mathbf{p} \cdot \pi}}{\partial \mathbf{p}} - \mathbf{p}.
\]

(29)

Inserting Eq. (29) into Eq. (25), we arrive at

\[
\langle n, \sigma, p | \pi | n', \sigma', p \rangle = m_0 \left( u_{n,\sigma}, p \left| \frac{\partial \mathcal{H}_{\mathbf{p} \cdot \pi}}{\partial \mathbf{p}} \right| u_{n',\sigma'}, p \right).
\]

(30)

A perturbation theoretical analysis of Eq. (30) up to first order in \( \mathbf{p} \) has already been performed previously\[16\], yielding

\[
\langle n, \sigma, p | \pi | n', \sigma', p \rangle = m_0 \left( u_{n,\sigma}, 0 \left| \frac{\partial \mathcal{H}_{\mathbf{p} \cdot \pi}}{\partial \mathbf{p}} \right| u_{n',\sigma'}, 0 \right).
\]

(31)

The \( \mathbf{p} \cdot \pi \) Hamiltonian \( \mathcal{H}_{\mathbf{p} \cdot \pi} \) describes the \( \mathbf{p} \)-dependent band dispersion in the crystal. In our system, this is identified with the kinetic energies of the electron in the conduction band \( \mathcal{H}_c \) and hole in the valence band \( \mathcal{H}_v \) respectively. Using the kinetic part of the Hamiltonian \( [1] \),

\[
T(p) = \mathcal{H}_c(p) - \mathcal{H}_v(p) = \frac{\gamma'_c}{2m_0} p^2 + \mathcal{H}_v(p),
\]

(32)

we can summarize all three cases in Eqs. (22)-(24) via

\[
\langle \Psi_{c',v} | \pi | \Psi_{c,v} \rangle
\]

(33)

\[
= m_0 \sum_p \phi_{c'}(p) \phi_{c'}^*(p) \langle c, \sigma_e, \mathbf{v}, \sigma_h | \partial_p T(p) | c', \sigma'_e, \mathbf{v}', \sigma'_h \rangle
\]

\[
= m_0 \int d^3r \psi_{c'}^\dagger(r) \langle c, \sigma_e, \mathbf{v}, \sigma_h | \partial_p T(p) | c', \sigma'_e, \mathbf{v}', \sigma'_h \rangle \psi_{c'}(r),
\]

(32)

where the matrix element is evaluated in the twelve-dimensional basis of electron-hole spin-states \( | c, \sigma_e, \mathbf{v}, \sigma_h \rangle \). The second line gives the equivalent expression in real space, where \( \psi_{c'}(r) \) is the real-space envelope function of the state \( | \Psi_{c',v} \rangle \). These states span the same Hilbert space as the basis states \( [1] \). Noting that only the kinetic energy terms in the Hamiltonian \( [1] \) contain the relative momentum operator \( \mathbf{p} \), we obtain the identity

\[
\pi = m_0 \frac{\partial}{\partial \mathbf{p}} H(p)
\]

(34)

valid for the one-exciton states considered in this paper.

Equation (34) is an operator identity in the one-exciton Hilbert space spanned, e.g., by the basis \( [11] \), and is valid for vanishing center-of-mass momentum \( \mathbf{p} \) and relative momentum \( \mathbf{p} \) much smaller than the extent of the Brillouin zone.

B. Numerical evaluation of the matrix elements

\( (\Pi | \pi | \Pi) \)

The computation of the dipole transition matrix elements \( M_{\Pi} \) in Eq. (18) requires one to evaluate the matrix elements \( (\Pi | \pi | \Pi) \) of the operator \( [34] \) in the basis \( [11] \).

From Eq. (34) we obtain

\[
\frac{\pi}{m_0} = \frac{\partial}{\partial \mathbf{p}} H(p) = \frac{\gamma'_c}{m_0} + \frac{\partial \mathcal{H}_v(p)}{\partial \mathbf{p}}.
\]

(35)

We focus on the component \( \pi_z \) for light polarized along the \( z \)-axis. The matrix elements for \( \pi_z \) in the basis \( [11] \) are derived in Appendix \( [A] \). The more difficult part is to evaluate the second term in Eq. (35). Instead of deriving the expression in detail here, we connect this problem to terms already calculated in Ref. [34]. They consider the Hamiltonian \( [1] \) in center-of-mass coordinates with with...
a nonvanishing center-of-mass momentum \( P = hK \) parallel to a given axis. Here, we are interested in the case \( P \parallel [001] \) related to the derivative with respect to \( p_z \). This means that we can set \( P = P_z \) in the following. Following Ref. 34, we expand the Hamiltonian in powers of \( P \)

\[
H(p, P) = H_0 + PH_1 + P^2 H_2. \tag{36}
\]

The center-of-mass transformation (3) is chosen in such a way that terms linear in \( P \) vanish without the corrections from the valence band. This means that the term \( H_1 \) arises solely from the kinetic energy \( H_k \) of the hole. More explicitly, we can write

\[
H_h(p_h) = -p + \alpha_h P
\]

\[
= \frac{p^2}{2m_h} + H_h(p) - \frac{\alpha_h P}{m_h} p_z + PH_1 + O(P^2),
\]

where \( \alpha_h = m_h / (m_h + m_e) \) is determined by the center-of-mass transformation. We first differentiate both sides with respect to \( P = P_z \) and evaluate at \( P = 0 \),

\[
\alpha_h \frac{\partial H_h}{\partial p_h z} (p_h = -p) = -\alpha_h \frac{\partial H_h}{\partial p_z} (-p) = -\frac{\alpha_h}{m_h} p_z + H_1.
\]

On the other hand, first setting \( P = 0 \) and differentiating with respect to \( p_z \) leads to

\[
\frac{\partial H_h}{\partial p_z} (-p) = \frac{p_z}{m_h} + \frac{\partial H_h(p)}{\partial p_z}.
\]

Comparing these results, we obtain the identity

\[
\frac{\partial H_h}{\partial p_z} (p) = -\frac{1}{\alpha_h} H_1 = -\frac{m_e}{m_h} \gamma_1 H_1.
\]

Inserted into Eq. (35), we finally find

\[
\gamma_1 = (p_z - m_e H_1),
\]

with 34

\[
H_1 = -\frac{1}{2\hbar^2 m_e} \left\{ 2\sqrt{\frac{5}{3}} \left[ P^{(1)} \times I^{(2)} \right]^{(1)}_{0} + 4\sqrt{\frac{2}{5}} \delta' \left[ P^{(1)} \times I^{(2)} \right]^{(3)}_{0} \right\}
\]

\[
-\frac{3\eta_1}{\gamma_1^2 h^2 m_e} \left\{ 2 \left[ P^{(1)} \times I^{(2)} \right]^{(1)}_{0} + 2\sqrt{\frac{5}{3}} \nu \left[ P^{(1)} \times D^{(2)} \right]^{(1)}_{0} + 4\sqrt{\frac{2}{5}} \tau \left[ P^{(1)} \times D^{(2)} \right]^{(3)}_{0} \right\},
\]

using the abbreviations

\[
D_k^{(2)} = \left[ I^{(1)} \times S_k^{(1)} \right]^{(2)}_{k}
\]

and

\[
\mu' = \frac{6\gamma_3 + 4\gamma_2}{5\gamma_1}, \quad \delta' = \gamma_3 + \gamma_2, \quad \nu = \frac{6\eta_3 + 4\eta_2}{5\eta_1}, \quad \theta = \eta_3 - \eta_2.
\]

A. Interseries absorption spectra

The transition matrix elements \( M_{fi} \) can be used to calculate interseries absorption spectra. The photoabsorption cross section \( \sigma_i \) from the initial state \( |\Psi_i\rangle \) at the spectral position \( E = h\omega_{ph} \) is given by 12, 35

\[
\sigma_i(\omega_{ph}) = \frac{4\pi\alpha h}{m_0^2 \omega_{ph}} \mathrm{Im} \sum_i E_i M_{fi} M_{fi}^*,
\]

with the fine-structure constant \( \alpha \) and \( h\omega_{ph} \approx E_i - E_f \). Note that in general, \( M_{fi} \neq M_{fi}^* \) for complex rotated states, and thus the numerator in Eq. (45) does not simplify to \( |M_{fi}|^2 \).

To avoid extremely narrow peaks for certain states, we phenomenologically model an additional linewidth caused by the coupling to phonons in the crystal. In a simplified model, the phonon-induced linewidth has a power-law dependency on the principal quantum number.

| \( E [\text{eV}] \) | \( 2P \times \text{R} \times M^2 \) | \( 3P \times \text{R} \times M^2 \) | \( 4P \times \text{R} \times M^2 \) | \( 5P \times \text{R} \times M^2 \) |
|-----------------|-----------------|-----------------|-----------------|-----------------|
| 2.28456         | -15.02 -19.56   | -1.36 1.11      | 0.11 -1.55      | 0.32 -0.71      |
| 2.28583         | 22.59 -0.60     | 3.92 -0.12      | 0.66 -0.03      | 0.23 -0.01      |
| 2.28895         | 366.60 1.17     | 15.29 0.41      | 0.17 0.03       | 0.00 0.00       |
| 2.28949         | 41.06 1.24      | 2.22 0.10       | 0.03 0.01       | 0.00 0.00       |
| 2.29283         | 3.00 7.36       | 13.42 -10.83    | 2.20 -2.18      | 1.87 -0.88      |
| 2.29367         | 43.45 -0.67     | 1.61 -0.11      | 1.25 -0.05      | 0.47 -0.02      |
| 2.29439         | 180.87 1.34     | 8.95 1.14       | 9.62 0.29       | 0.85 0.06       |
| 2.29494         | 46.04 -0.51     | 139.46 0.42     | 26.58 5.00      | 3.05 0.15       |
| 2.29522         | 0.82 0.08       | 16.55 0.80      | 3.92 0.09       | 0.34 0.03       |
| 2.29710         | 9.91 -0.04      | 30.51 -0.08     | 0.03 0.02       | 1.32 0.06       |
| 2.29776         | 19.31 -0.29     | 124.41 -0.22    | 62.81 -0.05     | 29.01 0.28      |
| 2.29845         | 33.28 0.18      | 9.36 0.55       | 21.16 -0.69     | 8.31 0.52       |
| 2.29864         | 16.24 0.73      | 1.87 0.02       | 4.49 -0.43      | 0.20 0.10       |
| 2.29932         | 10.08 -0.18     | 6.92 -0.11      | 2.06 -0.08      | 25.78 -0.20     |

IV. RESULTS AND DISCUSSION

In the following, we present our results for the dipole transition probabilities for two cases of interseries transitions. As parity is an exact quantum number, we separately discuss transitions first from odd parity to even parity states, and then from even parity to odd parity states. We choose a coordinate system where the \( x \)-, \( y \)-, and \( z \)-axes are parallel to \([001] \), \([010] \), and \([001] \) directions, respectively.
n as \[3, 30\]

\[
\gamma_{\text{ph}}(n) = \gamma_{\text{ph}} n^{-3}.
\]  

(46)

We estimate the parameter \(\gamma_{\text{ph}} = 56.4 \text{ meV}\) and assign to each resonance an effective quantum number \(n_{\text{eff}}\) based on the real part of its energy as outlined in Appendix [3]. The resulting linewidth shifts the imaginary part of the complex energy according to \(E_i \rightarrow E_i - i\gamma_{\text{ph}}/2\).

**B. Transitions from odd-parity yellow exciton states to even-parity green states**

Experimentally, the most easily accessible yellow exciton states are the odd-parity \(\Gamma_4^-\) \(P\)-states. As the interseries dipole transition flips the parity, the coupled states will be green even-parity states with \(S\)- and \(D\)-type envelopes. We now investigate two different scenarios. In the first, we select for the initial state the yellow \(P\)-exciton transforming like the basis state \(z\) of the irreducible representation \(\Gamma_4^-\) [37]. In the second scenario, we investigate the yellow \(P\)-exciton state transforming like the basis state \(y\). In both cases, the photon polarization is along the \(z\)-direction. From the product of the representations \[37\] \(\Gamma_4^- \otimes \Gamma_4^- = \Gamma_1^+ + \Gamma_3^+ + \Gamma_5^+ + \Gamma_6^+\) we can determine which transitions to green states are allowed in principle.

We begin with the yellow \(P\)-exciton and the photon both transforming according to the \(z\)-component of the \(\Gamma_4^-\) representation. This initial state can itself be excited using a one-photon absorption process with light polarized along the \(z\)-direction. Using the tables in Ref. [37], we can deduce that the corresponding green states transform according to \(\Gamma_1^+\) and the \(\psi_3^+\)-component of \(\Gamma_3^+\). In Fig. [2] we show interseries transition spectra in this configuration. We additionally list the results for a selection of states in Table [1].

Using the Rydberg energies of the yellow and green exciton series, we can estimate which green principal quantum number belongs to states with maximum overlap with a yellow exciton state with given principal quantum number. In the following, we use the values...
From a simple overlap argument, one would expect the transition strengths to be largest when initial and final state have comparable real-space extensions. As the linear extension of the excitons scales with the square of the state have comparable real-space extensions.

$$\frac{a_0^g}{a_0} \approx \frac{E_{\text{Ryd}}^g}{E_{\text{Ryd}}}.$$  \hspace{1cm} (47)

The Bohr radii $a_0^g$ and $a_0^i$ are related to the Rydberg energies by:

$$E_{\text{Ryd}}^g = 86.04 \text{ meV}$$ and $$E_{\text{Ryd}}^i = 150.4 \text{ meV}.$$
In the last two columns we show the transition matrix elements for the initial green states with the highest transition strengths move to higher energies, in accordance with Eq. (48).

Here, as well as in the following discussions, it is also evident in the configuration in Sec. IV C, where the initial state can be excited using a single-photon absorption process with light polarized along the $y$-
direction. Here, the corresponding green excitons transform like the $x$-component of $\Gamma_+^5$ and the $x$-$z$-component of $\Gamma_-^5$ [37]. In Fig. 3 we show a transition spectrum in this configuration. We additionally list the results for a selection of states in Table [III].

The strongest transition in Table [III] is from the $2P$ yellow exciton to the exciton state with energy $E = 2.25655 \text{eV}$, which is the lowest lying $2S$ state. Nevertheless, it is hardly visible in our simulated spectrum in Fig. 3 because of its much larger width as compared with the other states.

As the principal quantum number of the initial yellow state increases, the matrix elements here also become progressively weaker. The region of green states with the strongest transition from a given yellow state does not obey Eq. (48) as accurately as in the previous case, lying slightly lower energetically as expected. This could be related to the different spatial extensions of the addressed green states, in addition to the generally approximate character of the overlap argument.

C. Transitions from even parity yellow states to odd parity green states

We finally investigate transitions from yellow even-parity states to green odd-parity states. The former can be excited using two-photon absorption processes. For these transitions, we have to consider states with irreducible representations appearing in the tensor product $\Gamma_+^5 \otimes \Gamma_-^5 = \Gamma_2^- + \Gamma_3^+ + \Gamma_4^- + \Gamma_5^+$. In Fig. 4, we show spectra for transitions of this kind. As initial states, we chose excitons transforming according to the $xy$-component of the irreducible representation $\Gamma_5^-$. In Table [IV] we list the results for a selection of states. In Fig. 5 we additionally show the special case of the transitions where the initial state is the green $1S$ state, which is energetically placed among the yellow excitons.

The strongest transition in Table [IV] is from the $2S$ yellow exciton to the green $2P$ exciton state with energy $E = 2.28515 \text{eV}$. This is also the strongest transition we found among all configurations. This has to be balanced against the fact that the initial state is of even parity, which makes it inaccessible in one-photon transitions; it can, however, be excited using two-photon absorption.

We also investigated transitions from the $3D$ state, see panel (d) in Fig. 4. These seem to be substantially stronger than the transitions from the $3S$ states, but still weaker than those from the $1S$ and $2S$ excitons. Finally, there are several strong transitions starting from the green $1S$ exciton, but they are weaker than those from yellow $1S$ and $2S$ states.

V. CONCLUSION AND OUTLOOK

In this article, we have investigated interseries transitions between the yellow and green exciton series in the dipole approximation. We extended the calculations for the yellow-to-green interseries transitions performed in Ref. [10] by including the complex valence band structure. To properly take into account the associated coupling between the green exciton states and the yellow continuum, we used the complex-coordinate rotation method for the calculation of the green exciton resonances as described in Ref. [10].

We considered different choices for the initial state in the spectral range of the yellow series, concentrating mostly on the odd-parity $P$-states, which are most easily accessible in one-photon absorption experiments. We distinguished the cases where the photon that excites the initial exciton is polarized parallel to the photon affecting the interseries transition from the scenario in which they are orthogonally polarized. Additionally, we also calculated the probabilities for the transition from the even-parity yellow states to the odd-parity green states, with the special case where the initial state is the green $1S$-exciton.

The transition strengths are on the same order of magnitude in the different configurations, with those starting at an odd-parity yellow exciton being somewhat weaker than those starting at an even-parity yellow exciton. Of course, the experimental preparation of the latter is more difficult, as a two-photon excitation is required. In all cases, increasing the principal quantum number of the initial state shifts the range of excited green states to higher energies, with an overall weakening of the transition strengths in most cases.

In this work, we use the dipole approximation, which is valid if the wavelength of the light affecting the interseries transition is much larger than the extension of the involved excitons. As shown in Ref. [15], this condition breaks down for transitions between the yellow and green series starting at $n \gtrsim 15$ for counter-propagating pump and probe beams. Extending our investigations to this parameter range thus requires going beyond the dipole approximation. Furthermore, an extension of our
method to cover transitions between states of the yellow and blue series is relatively straightforward, but requires the implementation of the conduction band Hamiltonian including the $\Gamma_8$ band. Another possible route is to investigate the influence of an additional external field to fine tune the properties of the transitions.

Finally, one of the aims of our manuscript was to provide theoretical predictions which can help guide experimental investigations into the interseries transitions. While there has been some experimental work with respect to intraseries transitions within the yellow series [38, 39] and with respect to interseries transitions between the yellow and blue series [40], to the best of our knowledge, there have been no experimental studies into the yellow-to-green interseries transitions investigated in this manuscript yet. A comparison of our results with future experimental data is thus highly desirable.

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**Appendix A: The matrix element for $p_z$**

In the formalism of irreducible tensors, $p_z$ is given by

$$p_z = P^{(1)}_0.$$  \hfill (A1)

In the supplemental material of Ref. [34], Eq. (14) provides the matrix elements for the operator

$$P^{(1)}_0 \left( T^{(1)} \cdot S^{(1)}_h \right),$$  \hfill (A2)

which we can use here. Using the identity

$$T^{(1)} \cdot S^{(1)}_h = \frac{1}{2} \left( J^2 - I^2 - S^2_h \right) = \frac{2J(2J+2) - 11}{8},$$  \hfill (A3)

together with

$$\langle \Pi' | P^{(1)}_0 | \Pi \rangle = \frac{8}{2J(2J+2) - 11} \langle \Pi' | P^{(1)}_0 \left( T^{(1)} \cdot S^{(1)}_h \right) | \Pi \rangle.$$  \hfill (A4)

Here, $| \Pi \rangle$ and $| \Pi' \rangle$ denote basis states as given in Eq. (11).

**Appendix B: Phonon-induced linewidths**

In order to use Eq. (16), we need to determine the constant $\gamma_0^{ph}$. According to Ref. [7], the FWHM of the green $2P$-state at $T = 4K$ is $\gamma_0^{2P} = 17.7$ meV. In Ref. [10], the complex coordinate rotation method was used to calculate the complex energies of the odd-parity green excitons, and to determine the linewidths $\gamma_0^{cont}$ caused by the coupling of the green excitons to the yellow continuum. Here, we update this calculation by adding the Haken potential to the Hamiltonian, and find $\gamma_0^{2P} = 9.95$ meV for the green $2P$ state. We can thus estimate the phonon-induced linewidth of the $2P$ green exciton as

$$\gamma_{ph} (n=2) = \gamma_0^{2P} - \gamma_0^{cont} \approx 17.7 \text{ meV} - 9.95 \text{ meV} = 7.05 \text{ meV}$$  \hfill (B1)

leading to

$$\gamma_0^{ph} = 8 \times \gamma_{ph} (n=2) = 8 \times 7.05 \text{ meV} = 56.4 \text{ meV}.$$  \hfill (B2)

We associate to each resonance an effective quantum number $n_{eff}$ as a function of the real part of the resonance energy $E$,

$$n_{eff} = \sqrt{\frac{E_{Ryd}}{E_{gap} - E + \delta}}.$$  \hfill (B3)

The values $E_{Ryd} = 142$ meV, $E_{gap} = 2.30292$ eV and $\delta = 0.1$ were obtained by a phenomenological fit to the odd-parity green excitons in an updated version of the calculation in Ref. [10], where we included the Haken potential. Note that these values should not be taken as the literal Rydberg energy and quantum defect, as the inclusion of the Haken potential distorts the Rydberg spectrum.

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