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Self-consistent theory of the biexciton optical nonlinearity

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For the low-excitation regime a self-consistent set of equations for excitons (x), exciton molecules (m), and photons (γ) is derived from the electron-hole-photon Hamiltonian, taking the spin explicitly into account. In this approach the creation of m occurs via the Coulombic scattering of two x's of opposite internal spin structure. This process is resonant due to the polariton effect. Thus, the Coulombic matrix element for the process $x+x\rightarrow m$ governs the m formation, rather than the optical matrix element for the process $x+\gamma\rightarrow m$, as is usually assumed. At the same time, the scattering process $x+x\rightarrow m$ gives rise to a "true two-photon absorption" in the sense of Hopfield's concept of absorption in the polariton theory. Both for the 2γ absorption and for the x-m optical Stark effect, our theory gives measurable differences compared to the usual phenomenological approach. A Schrödinger equation for the m-wave function is derived which includes polariton effects.

I. INTRODUCTION

The concept of the giant two-photon (2γ) absorption of the excitonic molecule (m) has been introduced by Hanamura.[1] In this model the first step is the creation of an exciton (x) with momentum $\mathbf{p}$ by one-photon (γ) absorption and the second step is the optical conversion of this x to the m state $\mathbf{p}+\mathbf{k}$, where $\mathbf{k}$ is the wave number of the absorbed γ. The second transition has been attributed to the "giant" oscillator strength of the x-m optical transition. The strong enhancement of the 2γ absorption arises both from the nearly resonant first step and the large probability of the second one due to the spatial extension of m.

In this scheme, the 2γ absorption is determined by the momentum-dependent optical matrix element $M_1(\mathbf{p},\mathbf{k})$ of the process $x+\gamma\rightarrow m$, which is

$$M_1(\mathbf{p},\mathbf{k})=-i(\Omega_c/2)\Psi[(\mathbf{p}-\mathbf{k})/2]\ , \tag{1}$$

where $\Psi(\mathbf{l})$ is the m wave function which describes the relative motion of two bound x's. The polariton parameter Ω_c is defined in terms of the longitudinal-transverse (l-t) splitting ω_{lt} or by the dimensionless parameter β

$$\Omega_c^2=2\omega_{lt}\omega_t=(4\pi\beta/\epsilon_0)\omega_t^2\ . \tag{2}$$

Here, ω_t is the transverse x frequency and ϵ_0 is the background optical dielectric constant. The l-t splitting ω_{lt} determines the spectral width of the linear x-absorption line at low temperatures. The parameter $\Omega_c\gg\omega_{lt}$ characterizes the spectral range near the x line in which the corresponding nonlinear optical processes can be resonantly enhanced by the intermediate x state. This model of the 2γ-m absorption can be linked quite naturally with the polariton character of the propagation of the electromagnetic waves inside the crystal.

Biexcitons have been observed[2] first in the semiconductor CuCl which has a large direct energy gap. Due to the correspondingly small dielectric constant, the x and m binding energies of CuCl are very large. The concept of the giant 2γ-m absorption has been qualitatively supported by further experiments (see, e.g., Ref. 3). At present the interest in the optical properties of m's is renewed in connection with new high-precision techniques.[4-6] In particular, experimental investigations[7] of the Bose-Einstein condensation of m's are again under consideration.[8] Recently, the x-m optical Stark effect has been discovered[9] in CuCl. Therefore, we reinvestigate and refine the theory of the optical nonlinearities linked with the 2γ resonance of m.

In Sec. II, we derive for the low-excitation regime from the e-h-γ Hamiltonian a self-consistent x approximation for the description of the γ-x-m system. We show that the usual phenomenological approach, which treats γ, x, and m as independent unstructured bosons and introduces the optical matrix element $M_1(\mathbf{p},\mathbf{k})$ for the description of the 2γ-m absorption, is inconsistent with the basic e-h-γ picture. An important part of our theory is the consideration of the spin structure of the optically excited x's. We find an attractive, resonant Coulombic coupling of two x's with momenta $\mathbf{p}$ and $\mathbf{k}$ and appropriate spin structure to be responsible for the 2γ (two-polariton) creation of m with momentum $\mathbf{p}+\mathbf{k}$. The basic equations for the x-m optical Stark effect and the 2γ-m absorption of the new self-consistent approach and the phenomenological one based on M_1 are derived.

In Sec. III, we consider the low-intensity x-m optical Stark effect on the basis of the self-consistent x approach. The crucial feature of this treatment is the correct introduction of quantum-independent x's and m's in the presence of the polariton pump wave. The self-consistent description and the phenomenological one are compared.

In Sec. IV, the 2γ-m absorption coefficient is derived in the framework of the self-consistent model. It is shown that this approach and the phenomenological one yield different line shapes for the 2γ absorption spectra. High-resolution experiments should allow us to discriminate between the different line shapes predicted by both theories. The dependence of the 2γ-m absorption on the

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geometry of the exciting beams is calculated for both theories. Finally, we show that only the self-consistent description of the 2γ-m creation gives rise to a "true absorption" in the sense of Hopfield's polariton theory.

In Sec. V, we derive and analyze an alternative Schrödinger equation for the m-wave function which includes polariton effects. This equation yields the inverse radiative lifetime of an m. In order to make the description self-consistent this lifetime has to be put into the 2γ-m absorption coefficient. It is shown that for the m luminescence both approaches—the self-consistent and the phenomenological—yield the same results.

II. PHOTON-EXCITON-BIEXCITON SYSTEM IN DIRECT-GAP SEMICONDUCTORS

The optical properties of the electronic excitations of direct-gap semiconductors can be obtained from the following e-h-γ Hamiltonian:

$$H_{\mathrm{eh}\gamma}=H_{\mathrm{eh}}+H_{\gamma}+H_{\mathrm{eh}\text{-}\gamma} \;. \tag{3}$$

The e-h Hamiltonian H_{eh} is given by

$$H_{\mathrm{eh}}=\sum_{\mathbf{p}s}\hbar[\omega_{\mathbf{p}}^{e}a_{s;\mathbf{p}}^{\dagger}a_{s;\mathbf{p}}+\omega_{\mathbf{p}}^{h}b_{s;\mathbf{p}}^{\dagger}b_{s;\mathbf{p}}]+\tfrac{1}{2}\sum_{\substack{\mathbf{p},\mathbf{l},\mathbf{q}\neq 0\\ s,s'}}V_{\mathbf{q}}[a_{s;\mathbf{p}}^{\dagger}a_{s';\mathbf{l}}^{\dagger}a_{s';\mathbf{l}+\mathbf{q}}a_{s;\mathbf{p}-\mathbf{q}}+b_{s;\mathbf{p}}^{\dagger}b_{s';\mathbf{l}}^{\dagger}b_{s';\mathbf{l}+\mathbf{q}}b_{s;\mathbf{p}-\mathbf{q}}-2a_{s;\mathbf{p}}^{\dagger}b_{s';\mathbf{l}}^{\dagger}b_{s';\mathbf{l}+\mathbf{q}}a_{s;\mathbf{p}-\mathbf{q}}] \;. \tag{4}$$

Here,

$$\hbar\omega_{\mathbf{p}}^{e}=E_g+p^2/2m_e,\quad \hbar\omega_{\mathbf{p}}^{h}=p^2/2m_h \tag{5}$$

are the e,h energies with the corresponding operators $a_{s;\mathbf{p}}$ and $b_{s;\mathbf{p}}$, s is the spin projection, m_e and m_h are the effective e,h masses, E_g is the band gap, $V_{\mathbf{q}}=4\pi e^2/(V\epsilon_0 q^2)$ is the Fourier transform of the Coulomb potential, ϵ_0 is the static dielectric constant, and V is the crystal volume. We use $s=\pm 1$ for the e,h spin measured in units of $\hbar/2$.

The Hamiltonian H_{eh} describes free e-h pairs, and bound e-h complexes such as x's and m's. With this Hamiltonian, one can investigate the Coulombic interactions between various electronic excitations. These interactions give rise to nonlinear optical properties of the crystal.

The interaction Hamiltonian $H_{\mathrm{eh}\text{-}\gamma}$ describes the interband optical transitions. The conduction and valence bands are assumed to be formed by s states and by p states with $M_J=\pm\frac{3}{2}$, respectively. Then only e-h pairs with the total spin $S=0$ are dipole active in optical interband transitions. Photons γ with a given circular polarization excite e-h pairs with fixed spin directions, e.g., with e spin up and h spin down. Photons with opposite circular polarization excite e's with spin down and h's with spin up. The γ operators for the two circular polarizations $\sigma=1,2$ are called $\alpha_{\sigma;\mathbf{p}}$, respectively. The second part of the Hamiltonian (3) is

$$\begin{aligned}H_{\gamma}+H_{\mathrm{eh}\text{-}\gamma}=&\sum_{\sigma;\mathbf{p}}\hbar\omega_{\mathbf{p}}^{\gamma}\alpha_{\sigma;\mathbf{p}}^{\dagger}\alpha_{\sigma;\mathbf{p}}\\&+\sum_{\sigma;\mathbf{p},\mathbf{l}}[g_{\mathbf{l}}(\mathbf{p})\alpha_{\sigma;\mathbf{l}}a_{s;\mathbf{p}+\mathbf{l}}^{\dagger}b_{-s;-\mathbf{p}}^{\dagger}+\mathrm{H.c.}]\\&\qquad\text{where } s=-(-1)^{\sigma} \;,\end{aligned} \tag{6}$$

i.e., spin $s=+1$ for circular polarization $\sigma=1$ and spin $s=-1$ for $\sigma=2$. The γ frequency and the matrix element of the e-h optical transition are given by

$$\begin{aligned}&\omega_{\mathbf{p}}^{\gamma}=cp/\sqrt{\epsilon_0};\quad g_{\mathbf{l}}(\mathbf{p})=-\left[\frac{2\pi\hbar e^2}{Vcl}\right]^{1/2}v_{\mathrm{cv}}(\mathbf{p}) \;,\\&v_{\mathrm{cv}}(\mathbf{p})=-i\frac{\hbar}{m}\langle u_{\mathbf{p}+\mathbf{l},c}|\nabla|u_{-\mathbf{p},v}\rangle \;.\end{aligned} \tag{7}$$

$u_{\mathbf{p}+\mathbf{l},c}$ and $u_{-\mathbf{p},v}$ are the e and h Bloch functions, m and e are the mass and charge of a free electron, respectively.

For optical excitation with circularly polarized light, one has to introduce according to Eq. (6) two types of x's, both with total spin zero. In one x type the e spin is up, the h spin is down; in the other type, the e and h spin orientations are exchanged. The x operators $B_{\sigma;\mathbf{p}}$ are connected with the e,h Fermion pair operators by

$$B_{\sigma;j\mathbf{p}}=\frac{1}{\sqrt{V}}\sum_{\mathbf{l}}\phi_j(\mathbf{l})a_{s;\mathbf{l}+\alpha\mathbf{p}}b_{-s;-\mathbf{l}+\beta\mathbf{p}}$$

and

$$a_{s;\mathbf{l}+\alpha\mathbf{p}}b_{-s;-\mathbf{l}+\beta\mathbf{p}}=\frac{1}{\sqrt{V}}\sum_{j}B_{\sigma;j\mathbf{p}}\phi_j^{*}(\mathbf{l}) \;, \tag{8}$$

again with $s=-(-1)^{\sigma}$. The second relation follows from the completeness of the pair wave function $\phi_j(\mathbf{l})$ of state j in momentum space. The effective-mass ratios are given by $\alpha=m_e/M$ and $\beta=m_h/M$, where M is the total e-h mass $M=m_e+m_h$. The x operator fulfills boson commutators in the low-density regime. Corrections to the boson commutation relations are of the order $O(Na_x^3)$, where N is the pair concentration and a_x is the x Bohr radius. The m operator $A_{J\mathbf{K}}$ is given by

$$\begin{aligned}A_{J\mathbf{K}}=\frac{1}{2V^{3/2}}\sum_{\mathbf{p},\mathbf{l},\mathbf{q},s}&\Gamma_J(\mathbf{p},\mathbf{l},\mathbf{q})a_{s;\mathbf{l}+\alpha(\mathbf{p}+\mathbf{K}/2)}\\&\times b_{-s;-\mathbf{l}+\beta(\mathbf{p}+\mathbf{K}/2)}a_{-s;\mathbf{q}+\alpha(-\mathbf{p}+\mathbf{K}/2)}\\&\times b_{s;-\mathbf{q}+\beta(-\mathbf{p}+\mathbf{K}/2)} \;.\end{aligned} \tag{9}$$

Here, $\Gamma_J(\mathbf{p},\mathbf{l},\mathbf{q})$ is m-wave function normalized to unity. J is the complete quantum number of the internal m state with total spin zero. The bound m state with total spin zero can exist only if the two e's and the two h's have opposite spins, respectively. Thus the m-wave function has to be symmetric with respect to the permutation of two e's or h's:

$$\Gamma_J(\mathbf{p},\mathbf{l},\mathbf{q})=\Gamma_J(-\mathbf{p},\mathbf{q},\mathbf{l})=\Gamma_J(\mathbf{q}-\mathbf{l}+\beta\mathbf{p}-\alpha\mathbf{p},\beta\mathbf{q}+\alpha\mathbf{l}-2\alpha\beta\mathbf{p},\alpha\mathbf{q}+\beta\mathbf{l}+2\alpha\beta\mathbf{p})$$
$$=\Gamma_J(-\mathbf{q}+\mathbf{l}-\beta\mathbf{p}+\alpha\mathbf{p},\beta\mathbf{l}+\alpha\mathbf{q}+2\alpha\beta\mathbf{p},\alpha\mathbf{l}+\beta\mathbf{q}-2\alpha\beta\mathbf{p})\ . \qquad (10)$$

In order to investigate the dynamical properties of the γ-x-m system, one can derive from the e-h-γ Hamiltonian (3) a closed nonlinear set of Heisenberg equations for γ, x, and m. The resulting γ equation is

$$i\frac{\partial}{\partial t}\alpha_{\sigma;\mathbf{p}}=\omega_{\mathbf{p}}^{\gamma}\alpha_{\sigma;\mathbf{p}}+\sum_{\mathbf{l}}\frac{1}{\hbar}g_{\mathbf{p}}(\mathbf{l})b_{-s;-\mathbf{l}}a_{s;\mathbf{l}+\mathbf{p}}=\omega_{\mathbf{p}}^{\gamma}\alpha_{\sigma;\mathbf{p}}+\sum_{j}i\frac{\Omega_{cj}^{*}}{2}B_{\sigma;j\mathbf{p}}\ . \qquad (11)$$

The polariton parameter Ω_{cj} is

$$\Omega_{cj}=-\frac{2i}{\hbar}\sum_{\mathbf{l}}\frac{1}{\sqrt{V}}\phi_j(\mathbf{l})g_{\mathbf{p}}(\mathbf{l}-\beta\mathbf{p})\simeq 2\sqrt{2\pi}\frac{ev_{\mathrm{cv}}}{\sqrt{E_g}}\tilde{\phi}_j(\mathbf{R}=\mathbf{0})\ . \qquad (12)$$

Here, $\tilde{\phi}_{\mathrm{j}}(\mathbf{R})$ is the internal x-wave function in real space.

The Heisenberg equation for the dipole-active x's is given by

$$i\frac{\partial}{\partial t}B_{\sigma;j\mathbf{p}}=-\sum_{\mathbf{l}}\frac{1}{\hbar}g_{\mathbf{p}}(\mathbf{l}-\beta\mathbf{p})\frac{\phi_j(\mathbf{l})}{\sqrt{V}}\alpha_{\sigma;\mathbf{p}}$$
$$+\sum_{\mathbf{l}}\left[[\omega^{e}_{\mathbf{l}+\alpha\mathbf{p}}+\omega^{h}_{-\mathbf{l}+\beta\mathbf{p}}]\phi_j(\mathbf{l})-\sum_{\mathbf{l}'\neq\mathbf{l}}V_{\mathbf{l}-\mathbf{l}'}\phi_j(\mathbf{l}')\right]\frac{1}{\sqrt{V}}a_{s;\mathbf{l}+\alpha\mathbf{p}}b_{-s;-\mathbf{l}+\beta\mathbf{p}}$$
$$+\sum_{s';\mathbf{q},\mathbf{l},\mathbf{l}'\neq\mathbf{l}}V_{\mathbf{l}-\mathbf{l}'}\frac{\phi_j(\mathbf{l}')-\phi_j(\mathbf{l})}{\sqrt{V}}[a^{\dagger}_{s';\mathbf{l}'+\alpha\mathbf{q}}a_{s;\mathbf{l}'+\alpha\mathbf{p}}a_{s';\mathbf{l}+\alpha\mathbf{q}}b_{-s;-\mathbf{l}+\beta\mathbf{p}}+b^{\dagger}_{s';-\mathbf{l}'+\beta\mathbf{q}}b_{-s;-\mathbf{l}'+\beta\mathbf{p}}a_{s;\mathbf{l}+\alpha\mathbf{p}}b_{s';-\mathbf{l}+\beta\mathbf{q}}]$$
$$+\sum_{\mathbf{l},\mathbf{q}}\frac{1}{\hbar}\frac{\phi_j(\mathbf{l})}{\sqrt{V}}\alpha_{\sigma;\mathbf{q}}[g_{\mathbf{q}}(\mathbf{l}-\beta\mathbf{p})a^{\dagger}_{s;\mathbf{l}-\beta\mathbf{p}+\mathbf{q}}a_{s;\mathbf{l}+\alpha\mathbf{p}}+g_{\mathbf{q}}(\mathbf{l}+\alpha\mathbf{p}-\mathbf{q})b^{\dagger}_{-s;-\mathbf{l}-\alpha\mathbf{p}+\mathbf{q}}b_{-s;-\mathbf{l}+\beta\mathbf{p}}]\ . \qquad (13)$$

On the right-hand side, we recognize first the driving term due to the light field, next the free x motion, followed by Coulomb scattering terms and finally phase-space filling corrections to the driving term. The fixed spin s in Eqs. (11) and (13), is, as in Eq. (8), connected with the circular polarization $\sigma=1,2$ by $s=-(-1)^{\sigma}$.

In order to introduce the nonlinearities due to the x-x or x-m interactions on the right-hand side of Eq. (13), we limit ourselves in lowest order to the one-e and one-h subspaces.[10] The unit operator $\hat{I}$ in Fock space is

$$\hat{I}=\frac{1}{N!}\sum_{N,\mathbf{q}_i,s_i}|\mathbf{q}_1,s_1;\ldots;\mathbf{q}_N,s_N;e(h)\rangle_{A\,A}\langle\mathbf{q}_1,s_1;\ldots;\mathbf{q}_N,s_N;e(h)|$$
$$\simeq|0;e\rangle\langle 0;e|+\sum_{s_1,\mathbf{q}_1}a^{\dagger}_{s_1;\mathbf{q}_1}|0;e\rangle\langle 0;e|a_{s_1;\mathbf{q}_1}$$
$$=|0;h\rangle\langle 0;h|+\sum_{s_1,\mathbf{q}_1}b^{\dagger}_{s_1;\mathbf{q}_1}|0;h\rangle\langle 0;h|b_{s_1;\mathbf{q}_1}\ . \qquad (14)$$

Here $|\mathbf{q}_1,s_1;\ldots;\mathbf{q}_N;e(h)\rangle_A$ is the antisymmetric state of the crystal with N e or h. The latter approximations hold in the x regime where $Na_x^3<1$. One has to insert this unit operator in the Coulomb scattering terms and the phase-space filling terms of Eq. (13) to generate the x (ab) and m ($abab$) coupling of the e and h operators. The parts connected with the first term $|0;e(h)\rangle\langle 0;e(h)|$ of the approximation (14) are zero for any x and m states, i.e., in the absence of unpaired e and h. Thus these parts do not contribute to the x and m nonlinearities. The first nonvanishing contributions to these nonlinearities come from the second terms in Eq. (14). With this procedure, the Coulomb scattering and the phase-space filling terms of Eq. (13) are expressed uniquely in terms of e-h pair operators:

$$[a^{\dagger}_{s';\mathbf{l}'+\alpha\mathbf{q}}a_{s;\mathbf{l}'+\alpha\mathbf{p}}a_{s';\mathbf{l}+\alpha\mathbf{q}}b_{-s;-\mathbf{l}+\beta\mathbf{p}}+b^{\dagger}_{s';-\mathbf{l}'+\beta\mathbf{q}}b_{-s;-\mathbf{l}'+\beta\mathbf{p}}a_{s;\mathbf{l}+\alpha\mathbf{p}}b_{s';-\mathbf{l}+\beta\mathbf{q}}]$$
$$=\sum_{\mathbf{q}_1,s_1}[a^{\dagger}_{s';\mathbf{l}'+\alpha\mathbf{q}}b^{\dagger}_{s_1;\mathbf{q}_1}b_{s_1;\mathbf{q}_1}a_{s;\mathbf{l}'+\alpha\mathbf{p}}a_{s';\mathbf{l}+\alpha\mathbf{q}}b_{-s;-\mathbf{l}+\beta\mathbf{p}}+b^{\dagger}_{s';-\mathbf{l}'+\beta\mathbf{q}}a^{\dagger}_{s_1;\mathbf{q}_1}a_{s_1;\mathbf{q}_1}b_{-s;-\mathbf{l}'+\beta\mathbf{p}}a_{s;\mathbf{l}+\alpha\mathbf{p}}b_{s';-\mathbf{l}+\beta\mathbf{q}}]\ ,$$
$$a^{\dagger}_{s;\mathbf{l}-\beta\mathbf{p}+\mathbf{q}}a_{s;\mathbf{l}+\alpha\mathbf{p}}=\sum_{\mathbf{q}_1,s_1}a^{\dagger}_{s;\mathbf{l}-\beta\mathbf{p}+\mathbf{q}}b^{\dagger}_{s_1;\mathbf{q}_1}b_{s_1;\mathbf{q}_1}a_{s;\mathbf{l}+\alpha\mathbf{p}}\ , \qquad (15)$$
$$b^{\dagger}_{-s;-\mathbf{l}-\alpha\mathbf{p}+\mathbf{q}}b_{-s;-\mathbf{l}+\beta\mathbf{p}}=\sum_{\mathbf{q}_1,s_1}b^{\dagger}_{-s;-\mathbf{l}-\alpha\mathbf{p}+\mathbf{q}}a^{\dagger}_{s_1;\mathbf{q}_1}a_{s_1;\mathbf{q}_1}b_{-s;-\mathbf{l}+\beta\mathbf{p}}\ .$$

This procedure is valid up to order $(Na_x^3)^2$. The x and m coupling does not depend on the position where the unit operator $\hat{1}$ is inserted. For definiteness, we use the normal order of the operators in Eq. (15).

Now the x and m operators can be introduced. Equation (13) takes the form

$$i\frac{\partial}{\partial t}B_{\sigma;j\mathbf{p}}=\omega_{j\mathbf{p}}^{x}B_{\sigma;j\mathbf{p}}-i\frac{\Omega_{cj}}{2}\alpha_{\sigma;\mathbf{p}}+\mathbf{J}_{\sigma,\sigma}^{x-x}+\mathbf{J}_{\sigma\neq\sigma'}^{x,x\to m}\ , \tag{16}$$

where $\hbar\omega_{j\mathbf{p}}^{x}=E_g-\epsilon_j^x+\hbar^2p^2/2M$ is the x frequency of state j, and ϵ_j^x is the corresponding binding energy.

The term $J_{\sigma,\sigma}^{x,x}$ describes the time development due to the repulsive scattering between two x's of the same spin configurations σ, while $J_{\sigma,\sigma'\neq\sigma}^{x,x\to m}$ is due to the attractive interaction between two x's with opposite spin configuration $\sigma\neq\sigma'$ which gives rise to m formation. The repulsive x-x interaction is given by

$$\begin{aligned}\mathbf{J}_{\sigma,\sigma}^{x,x}=-\sum_{\substack{\mathbf{q},\boldsymbol{l},\boldsymbol{l}'\neq\boldsymbol{l}\\ i_1,i_2,i_3}}\frac{V_{l-l'}}{V^2}\{&\phi_j(\boldsymbol{l})\phi_{i_1}^*(\boldsymbol{l})[\phi_{i_2}^*(\boldsymbol{l}'-\beta\mathbf{p}+\beta\mathbf{q})\phi_{i_3}(\boldsymbol{l}'-\beta\mathbf{p}+\beta\mathbf{q})+\phi_{i_2}^*(\boldsymbol{l}'+\alpha\mathbf{p}-\alpha\mathbf{q})\phi_{i_3}(\boldsymbol{l}'+\alpha\mathbf{p}-\alpha\mathbf{q})]\\ &-\phi_j(\boldsymbol{l})\phi_{i_1}^*(\boldsymbol{l}')[\phi_{i_2}^*(\boldsymbol{l}-\beta\mathbf{p}+\beta\mathbf{q})\phi_{i_3}(\boldsymbol{l}-\beta\mathbf{p}+\beta\mathbf{q})\\ &+\phi_{i_2}^*(\boldsymbol{l}+\alpha\mathbf{p}-\alpha\mathbf{q})\phi_{i_3}(\boldsymbol{l}+\alpha\mathbf{p}-\alpha\mathbf{q})]\}B_{\sigma;i_3\mathbf{q}}^{\dagger}B_{\sigma;i_2\mathbf{q}}B_{\sigma;i_1\mathbf{p}}\\ +\sum_{\substack{\boldsymbol{l},\mathbf{q}\\ i_1,i_2}}\frac{g_{\mathbf{p}}(\boldsymbol{l}-\beta\mathbf{p})\phi_j(\boldsymbol{l})}{\hbar V^{3/2}}&[\phi_{i_1}(\boldsymbol{l}-\beta\mathbf{p}+\beta\mathbf{q})\phi_{i_2}^*(\boldsymbol{l}-\beta\mathbf{p}+\beta\mathbf{q})\\ &+\phi_{i_1}(\boldsymbol{l}+\alpha\mathbf{p}-\alpha\mathbf{q})\phi_{i_2}^*(\boldsymbol{l}+\alpha\mathbf{p}-\alpha\mathbf{q})]B_{\sigma;i_2\mathbf{q}}^{\dagger}B_{\sigma;i_1\mathbf{q}}\alpha_{\sigma;\mathbf{p}}\ .\end{aligned} \tag{17}$$

If a coherent electromagnetic pump wave with circular polarization excites only a coherent x amplitude of the ground state $j=1$ with momentum $\mathbf{p}$, and if scattering into higher states $j>1$ is neglected, Eqs. (16) and (17) can be reduced to

$$\begin{aligned}i\frac{\partial}{\partial t}\langle B_{\sigma;1,\mathbf{p}}\rangle=&\left[\omega_{1\mathbf{p}}^{x}+\sum_{\boldsymbol{l},\boldsymbol{l}'\neq\boldsymbol{l}}\frac{1}{V^2}V_{l-l'}|\phi_1(\boldsymbol{l})|^2[\phi_1(\boldsymbol{l})-\phi_1(\boldsymbol{l}')]\phi_1^*(\boldsymbol{l})|\langle B_{\sigma;1,\mathbf{p}}\rangle|^2\right]\langle B_{\sigma;1,\mathbf{p}}\rangle\\ &-\sum_{\boldsymbol{l}}\frac{1}{\hbar}g_{\mathbf{p}}(\boldsymbol{l}-\beta\mathbf{p})\frac{\phi_1(\boldsymbol{l})}{\sqrt{V}}[1-|\phi_1(\boldsymbol{l})|^2|\langle B_{\sigma;1,\mathbf{p}}\rangle|^2]\langle\alpha_{\sigma;\mathbf{p}}\rangle\ .\end{aligned} \tag{18}$$

This equation together with Eq. (11) are the basis of the x optical Stark effect in the so-called weak-field limit.[11,12] Both dynamical x level shift due to the x-x interaction and the saturation of the x-γ optical transition are contained in Eq. (18).

The attractive x-x interaction $J_{\sigma,\sigma\neq\sigma}^{x,x\to m}$ can be written in the form

$$\begin{aligned}J_{\sigma,\sigma\neq\sigma}^{x,x\to m}=\tfrac{1}{8}\sum_{\substack{\mathbf{k},\mathbf{q},\mathbf{q}'\\(\mathbf{p}+\mathbf{k}=\mathbf{q}+\mathbf{q}')\\ i_1,i_2,i_3}}&\frac{1}{V^2\sqrt{V}}[W_{\sigma,\sigma'\neq\sigma}(j\mathbf{p},i_1\mathbf{k},i_2\mathbf{q},i_3\mathbf{q}')+W_{\sigma,\sigma'\neq\sigma}(i_1\mathbf{k},j\mathbf{p},i_2\mathbf{q},i_3\mathbf{q}')]\\ &\times\sum_{\boldsymbol{l},\boldsymbol{l}'}\phi_{i_2}(\boldsymbol{l})\phi_{i_3}(\boldsymbol{l}')\Gamma_J^*\left[\frac{\mathbf{q}-\mathbf{q}'}{2},\boldsymbol{l},\boldsymbol{l}'\right]B_{\sigma'\neq\sigma;i_1\mathbf{k}}^{\dagger}A_{J,\mathbf{p}+\mathbf{k}}\ .\end{aligned} \tag{19}$$

Here $W_{\sigma,\sigma'\neq\sigma}(j_1\mathbf{p}_1,j_2\mathbf{p}_2,j_3\mathbf{p}_3,j_4\mathbf{p}_4)$ is the Fourier transform of the x-x interaction potential

$$W_{\sigma,\sigma'\neq\sigma}(j_1\mathbf{p}_1,j_2\mathbf{p}_2,j_3\mathbf{p}_3,j_4\mathbf{p}_4)=W_d(j_1\mathbf{p}_1,j_2\mathbf{p}_2,j_3\mathbf{p}_3,j_4\mathbf{p}_4)-W_{\mathrm{ex}}(j_1\mathbf{p}_1,j_2\mathbf{p}_2,j_3\mathbf{p}_3,j_4\mathbf{p}_4)\ . \tag{20}$$

The direct interaction potential is

$$\begin{aligned}W_d(j_1\mathbf{p}_1,j_2\mathbf{p}_2,j_3\mathbf{p}_3,j_4\mathbf{p}_4)=\int d^3R_e\,d^3R_h\,d^3R_e'\,d^3R_h'\,&\Phi_{j_1\mathbf{p}_1}^*(\mathbf{R}_e,\mathbf{R}_h)\Phi_{j_2\mathbf{p}_2}^*(\mathbf{R}_e',\mathbf{R}_h')\\ &\times\frac{e^2}{\bar{\epsilon}_0}\left[\frac{1}{|\mathbf{R}_e-\mathbf{R}_e'|}+\frac{1}{|\mathbf{R}_h-\mathbf{R}_h'|}-\frac{1}{|\mathbf{R}_e-\mathbf{R}_h'|}-\frac{1}{|\mathbf{R}_h-\mathbf{R}_e'|}\right]\\ &\times\Phi_{j_3\mathbf{p}_3}(\mathbf{R}_e,\mathbf{R}_h)\Phi_{j_4\mathbf{p}_4}(\mathbf{R}_e',\mathbf{R}_h')\ .\end{aligned} \tag{21}$$

The exchange interaction potential is

$$W_{\mathrm{ex}}(j_1\mathbf{p}_1,j_2\mathbf{p}_2,j_3\mathbf{p}_3,j_4\mathbf{p}_4)=\int d^3R_e d^3R_h d^3R_e' d^3R_h' \Phi^*_{j_1\mathbf{p}_1}(\mathbf{R}_e,\mathbf{R}_h)\Phi^*_{j_2\mathbf{p}_2}(\mathbf{R}_e',\mathbf{R}_h')$$
$$\times\frac{e^2}{\tilde{\epsilon}_0}\left[\frac{1}{|\mathbf{R}_e-\mathbf{R}_e'|}+\frac{1}{|\mathbf{R}_h-\mathbf{R}_h'|}-\frac{1}{|\mathbf{R}_e-\mathbf{R}_h'|}-\frac{1}{|\mathbf{R}_h-\mathbf{R}_e'|}\right]$$
$$\times\Phi_{j_3\mathbf{p}_3}(\mathbf{R}_e',\mathbf{R}_h)\Phi_{j_4\mathbf{p}_4}(\mathbf{R}_e,\mathbf{R}_h')\ , \tag{22}$$

which is obtained from W_d by interchanging the positions of the two e's in the last two wave functions. The complete x-wave function $\Phi_{j\mathbf{p}}(\mathbf{R}_e,\mathbf{R}_h)$ of state j with translation momentum $\mathbf{p}$ is given by

$$\Phi_{j\mathbf{p}}(\mathbf{R}_e,\mathbf{R}_h)=\frac{1}{\sqrt{V}}\exp[i\mathbf{p}\cdot(\alpha\mathbf{R}_e+\beta\mathbf{R}_h)]\tilde{\phi}_j(\mathbf{R}_e-\mathbf{R}_h)\ . \tag{23}$$

In the derivation of Eq. (19), the symmetry properties (10) of the m-wave function $\Gamma_J(\mathbf{p},\boldsymbol{l},\mathbf{q})$ plays an important role. If the complete m-wave function is symmetric in the spin variables, i.e., when spins of the both e's (and h's) have the same directions, the function (20) $W_{\sigma,\sigma'\neq\sigma}=W_d-W_{\mathrm{ex}}$ has to be replaced in Eq. (19) by $W_{\sigma\sigma}=W_d+W_{\mathrm{ex}}$. This change leads to the absence of the bound m state with this spin structure.

According to Eq. (16), one can divide the x nonlinearity with the terms $J^{x,x}_{\sigma,\sigma}$ and $J^{x,x\to m}_{\sigma,\sigma\neq\sigma}$ in a natural manner. In a "pump-probe" experiment the first term which is responsible for the x optical Stark effect plays a role only if the pump and probe beams have the same circular polarization. For opposite circular polarizations of both beams, the transitions to the m state as well as the x-m optical Stark effect can be realized. The description of these phenomena is directly connected with the term $J^{x,x\to m}_{\sigma,\sigma'\neq\sigma}$. This result is very similar to the conclusions obtained in a recent experimental study.[13]

In the weak-excitation regime $Na_x^3\ll 1$, one can derive directly from the e,h Hamiltonian a Hamiltonian for interacting x's,[10,14–17] either with the boson formalism of Usui[18] or with the much simpler unit operator technique:

$$H_x=\sum_{\sigma;\mathbf{p},j}\hbar\omega^x_{j\mathbf{p}}B^\dagger_{\sigma;j\mathbf{p}}B_{\sigma;j\mathbf{p}}$$
$$+\frac{1}{2}\sum_{\sigma,\sigma';\mathbf{p}_i,j_i}W_{\sigma,\sigma'}(j_1\mathbf{p}_1,j_2\mathbf{p}_2,j_3\mathbf{p}_3,j_4\mathbf{p}_4)$$
$$\times B^\dagger_{\sigma;j_1\mathbf{p}_1}B^\dagger_{\sigma';j_2\mathbf{p}_2}B_{\sigma';j_3\mathbf{p}_3}B_{\sigma;j_4\mathbf{p}_4}\ . \tag{24}$$

For the description of the x system, this Hamiltonian approximates the original e-h Hamiltonian (4) up to order $(Na_x^3)^2$. The polariton effects can be incorporated in the x representation (24) by rewriting $H_\gamma+H_{\mathrm{eh}\text{-}\gamma}$ in terms of x operators,

$$H_\gamma+H_{\mathrm{eh}\text{-}\gamma}=\sum_{\sigma;\mathbf{p}}\hbar\omega^\gamma_p\alpha^\dagger_{\sigma;\mathbf{p}}\alpha_{\sigma;\mathbf{p}}$$
$$+\sum_{\sigma;\mathbf{p},j}i\hbar\left[\frac{\Omega^*_{cj}}{2}\alpha^\dagger_{\sigma;\mathbf{p}}B_{\sigma;j\mathbf{p}}-\frac{\Omega_{cj}}{2}B^\dagger_{\sigma;j\mathbf{p}}\alpha_{\sigma;\mathbf{p}}\right]\ . \tag{25}$$

Equation (16) for the x's also follows directly from the boson Hamiltonian (24) and (25).

The Heisenberg equation for the m operator defined in Eq. (9) is given by

$$i\frac{\partial}{\partial t}A_{J\mathbf{K}}=\Omega^m_{J\mathbf{K}}A_{J\mathbf{K}}+\sum_{\substack{\mathbf{p},\boldsymbol{l},\mathbf{q}\\ i,\sigma}}\frac{g_{-\mathbf{p}+\mathbf{K}/2}(\mathbf{q})\phi^*_i(\boldsymbol{l})}{\hbar\sqrt{V}}\Gamma_J(\mathbf{p},\boldsymbol{l},\mathbf{q})$$
$$\times\alpha_{\sigma;-\mathbf{p}+\mathbf{K}/2}B_{\sigma'\neq\sigma;\mathbf{p}+\mathbf{K}/2}\ . \tag{26}$$

Here $\Omega^m_{J\mathbf{K}}$ is the m energy. The internal m-wave function $\Gamma_J(\mathbf{p},\mathbf{l},\mathbf{q})$ is determined by the following Schrödinger equation:

$$\Omega^m_{J\mathbf{K}}\Gamma_J(\mathbf{p},\boldsymbol{l},\mathbf{q})=[\omega^e_{\mathbf{q}-\alpha\mathbf{p}+\alpha\mathbf{K}/2}+\omega^e_{\boldsymbol{l}+\alpha\mathbf{p}+\alpha\mathbf{K}/2}+\omega^h_{-\mathbf{q}-\beta\mathbf{p}+\beta\mathbf{K}/2}+\omega^h_{-\boldsymbol{l}+\beta\mathbf{p}+\beta\mathbf{K}/2}]\Gamma_J(\mathbf{p},\boldsymbol{l},\mathbf{q})$$
$$-\sum_{\mathbf{q}'}V_{\mathbf{q}'}[\Gamma_J(\mathbf{p},\boldsymbol{l},\mathbf{q}+\mathbf{q}')+\Gamma_J(\mathbf{p},\boldsymbol{l}+\mathbf{q}',\mathbf{q})]$$
$$+\sum_{\mathbf{q}'}V_{\mathbf{q}'}[\Gamma_J(\mathbf{p}-\mathbf{q}',\boldsymbol{l}+\alpha\mathbf{q}',\mathbf{q}+\beta\mathbf{q}')+\Gamma_J(\mathbf{p}-\mathbf{q}',\boldsymbol{l}-\beta\mathbf{q}',\mathbf{q}-\alpha\mathbf{q}')$$
$$-\Gamma_J(\mathbf{p}-\mathbf{q}',\boldsymbol{l}+\alpha\mathbf{q}',\mathbf{q}-\alpha\mathbf{q}')-\Gamma_J(\mathbf{p}-\mathbf{q}',\boldsymbol{l}-\beta\mathbf{q}',\mathbf{q}+\beta q')]\ . \tag{27}$$

The first terms on the right-hand side of Eq. (27) describe the energies of the two x's in the m, whereas the last terms are due to their Coulombic interaction. The m Schrödinger equation (27) can again be obtained on the basis of the x approach (24).[15,16]

Equations (11), (16) (in which the term $J^{xx\to m}_{\sigma,\sigma'\neq\sigma}$ is kept but the term $J^{x,x}_{\sigma,\sigma}$ is omitted), and (26) form a closed nonlinear set for the γ-x-m system. Unfortunately, this set is rather complicated for analytical considerations. For the following analysis we introduce therefore further simplifications.

For the investigations of two resonantly interacting polariton waves due to the 2γ-m creation, one can use the x approximation which holds for $(N_1+N_2)a_x^3<1$. Here, $N_{i=1,2}\propto I_{i=1,2}$ are the concentrations of the x components of two interacting polaritons waves $(\mathbf{p},\omega)$ and $(\mathbf{k},\omega_{\mathbf{k}})$ with intensities I_1 and I_2, respectively. This assumption may be used because the *m* nonlinearities (e.g., the *x*-*m* optical Stark effect and the 2γ-*m* absorption) arise already at a lower excitation level $(N_1+N_2)a_m^3 \le 1$. Here, a_m is the *m* radius in the ground state.

The *x* theory can be simplified further if one considers only *x*'s in the ground state. This assumption is valid in the immediate spectral vicinity of the corresponding *x* absorption peak and holds, e.g., in the experimental investigations[2–9] of the *m* nonlinearity in the CuCl and to a lesser degree in CdS crystals, provided the light pulses are not too short and therefore spectrally not too broad. Furthermore, if the *m* binding energy is considerably smaller than the *x* binding energy, as for CuCl and CdS, one can use the following adiabatic approximation[15,16] for the *m*-wave function:

$$\begin{aligned}\Gamma_J(\mathbf{p},\boldsymbol{l},\mathbf{q}) =& \Psi_J(\mathbf{p})\phi_1(\boldsymbol{l})\phi_1(\mathbf{q})+\Psi_J(-\mathbf{p})\phi_1(\mathbf{q})\phi_1(\boldsymbol{l})\\ &-\Psi_J(\mathbf{q}-\boldsymbol{l}+\beta\mathbf{p}-\alpha\mathbf{p})\phi_1(\beta\mathbf{q}+\alpha\boldsymbol{l}-2\alpha\beta\mathbf{p})\phi_1(\alpha\mathbf{q}+\beta\boldsymbol{l}+2\alpha\beta\mathbf{p})\\ &-\Psi_J(-\mathbf{q}+\boldsymbol{l}-\beta\mathbf{p}+\alpha\mathbf{p})\phi_1(\beta\boldsymbol{l}+\alpha\mathbf{q}+2\alpha\beta\mathbf{p})\phi_1(\alpha\boldsymbol{l}+\beta\mathbf{q}-2\alpha\beta\mathbf{p})\ . \qquad (28)\end{aligned}$$

This approximation satisfies the condition (10) and allows us to describe the *m* state by means of the wave function $\Psi_J(\mathbf{p})$ for the relative motion of the two bound *x*'s. Such approximations have been used in the many theoretical investigations[19–23] of the *m* nonlinearity.

Thus if only the *x* ground state is considered, the *x* Hamiltonian (24) and (25) simplifies to[24,25]

$$\begin{aligned}H =& \hbar\sum_{\mathbf{p},\sigma}\left[\omega_{\mathbf{p}}^{x}B_{\sigma;\mathbf{p}}^{\dagger}B_{\sigma;\mathbf{p}}+\omega_{\mathbf{p}}^{\gamma}\alpha_{\sigma;\mathbf{p}}^{\dagger}\alpha_{\sigma;\mathbf{p}}\right.\\ &\left.+\left[\frac{i\Omega_c}{2}\alpha_{\sigma;\mathbf{p}}^{\dagger}B_{\sigma;\mathbf{p}}+\text{H.c.}\right]\right]\\ &+\frac{1}{2}\sum_{\substack{\mathbf{p},\boldsymbol{l},\mathbf{q}\\ \sigma,\sigma'}}W_{\sigma\sigma'}(\mathbf{q})B_{\sigma;\mathbf{p}}^{\dagger}B_{\sigma';\boldsymbol{l}}^{\dagger}B_{\sigma';\boldsymbol{l}+\mathbf{q}}B_{\sigma;\mathbf{p}-\mathbf{q}}\ , \qquad (29)\end{aligned}$$

where $B_{\sigma;j=1,\mathbf{p}}\equiv B_{\sigma;\mathbf{p}}$, and $\omega_{\mathbf{p}}^{x}=\omega_t+\hbar p^2/2M$. The polariton parameter Ω_c is given by Eq. (12), with $j=1$. Now the potential $W_{\sigma\sigma'}(\mathbf{q})$ of the *x*-*x* interaction depends only on the transferred momentum $\mathbf{q}$ (Ref. 15) and is assumed to be real. Although the *x*'s are treated here as elementary particles, the potential $W_{\sigma\sigma'}(\mathbf{q})$ (for explicit expressions, see Ref. 15) still has the important property that it is repulsive for two *x*'s with $\sigma=\sigma'$ and attractive for two *x*'s with $\sigma\neq\sigma'$ $(W_{12}=W_{21}<0)$, which leads to *m* formation.

In the considered approximations, the *m* operator $A_{J\mathbf{K}}$ is given by

$$\begin{aligned}A_{J\mathbf{K}} &= \frac{1}{2\sqrt{V}}\sum_{\sigma;\boldsymbol{l}}\Psi_J(\boldsymbol{l})B_{\sigma;\boldsymbol{l}+\mathbf{K}/2}B_{\sigma'\neq\sigma;-\boldsymbol{l}+\mathbf{K}/2}\\ &= \frac{1}{\sqrt{V}}\sum_{\boldsymbol{l}}\Psi_J(\boldsymbol{l})B_{1;\boldsymbol{l}+\mathbf{K}/2}B_{2;-\boldsymbol{l}+\mathbf{K}/2}\ . \qquad (30)\end{aligned}$$

The *x* Hamiltonian (29) does not explicitly contain the *m*'s, i.e., the operators $A_{J\mathbf{K}}$. The complete set of *m* eigenfunctions $\Psi_J(\boldsymbol{l})$ for all bound and ionized states can be found together with the corresponding *m* energies $\Omega_{J\mathbf{K}}^{m}$ from the following Schrödinger equation in the momentum representation:

$$\begin{aligned}&\frac{\hbar^2 l^2}{M}\Psi_J(\boldsymbol{l})+\sum_{\boldsymbol{l}'}W_{12}(\boldsymbol{l}-\boldsymbol{l}')\Psi_J(\boldsymbol{l}')\\ &\quad=\left[\Omega_{J\mathbf{K}}^{m}-2\omega_t-\frac{\hbar^2K^2}{4M}\right]\Psi_J(\boldsymbol{l})=-\epsilon_J^{m}\Psi_J(\boldsymbol{l})\ . \qquad (31)\end{aligned}$$

Multiplying Eq. (30) by $\Psi_J^*(\boldsymbol{l})$, we obtain, with the completeness of $\{\Psi_J(\boldsymbol{l})\}$,

$$\begin{aligned}B_{\sigma;\boldsymbol{l}+\mathbf{K}/2}B_{\sigma'\neq\sigma;-\boldsymbol{l}+\mathbf{K}/2} &= \frac{1}{\sqrt{V}}\sum_{J}\Psi_J^*(\boldsymbol{l})A_{J\mathbf{K}}\\ &\simeq \frac{1}{\sqrt{V}}\Psi^*(\boldsymbol{l})A_{\mathbf{K}}\ , \qquad (32)\end{aligned}$$

where the last approximation is valid only under resonant condition for the *m* ground state.

In the considered case $(N_1+N_2)a_m^3\le 1$, the Heisenberg equations for the γ, *x*, and *m* operators are

$$i\frac{\partial}{\partial t}\alpha_{\sigma;\mathbf{p}}=\omega_{\mathbf{p}}^{\gamma}\alpha_{\sigma;\mathbf{p}}+i\frac{\Omega_c}{2}B_{\sigma;\mathbf{p}}\ , \qquad (33)$$

$$i\frac{\partial}{\partial t}B_{\sigma;\mathbf{p}}=\omega_{\mathbf{p}}^{x}B_{\sigma;\mathbf{p}}-i\frac{\Omega_c}{2}\alpha_{\sigma;\mathbf{p}}+\sum_{\boldsymbol{l}}\frac{1}{\sqrt{V}}M_2^*(\mathbf{p},\boldsymbol{l})B_{\sigma'\neq\sigma;\boldsymbol{l}}^{\dagger}A_{\mathbf{p}+\boldsymbol{l}}\ , \qquad (34)$$

$$\begin{aligned}i\frac{\partial}{\partial t}A_{\mathbf{K}} =& \sum_{\boldsymbol{l}}\left\{[\omega_{\boldsymbol{l}+\mathbf{K}/2}^{x}+\omega_{-\boldsymbol{l}+\mathbf{K}/2}^{x}]\Psi(\boldsymbol{l})+\sum_{\boldsymbol{l}'}W_{12}(\boldsymbol{l}-\boldsymbol{l}')\Psi(\boldsymbol{l}')\right\}\frac{1}{\sqrt{V}}B_{1;\boldsymbol{l}+\mathbf{K}/2}B_{2;-\boldsymbol{l}+\mathbf{K}/2}\\ &+\sum_{\sigma;\boldsymbol{l}}\frac{1}{\sqrt{V}}M_1(\boldsymbol{l}+\mathbf{K}/2,\boldsymbol{l}-\mathbf{K}/2)\alpha_{\sigma;\boldsymbol{l}+\mathbf{K}/2}B_{\sigma'\neq\sigma;-\boldsymbol{l}+\mathbf{K}/2}\ , \qquad (35)\end{aligned}$$

where the matrix element $M_2(\mathbf{p},\mathbf{l})$ is given by

$$M_2(\mathbf{p},\mathbf{l})=\Delta[(\mathbf{p}-\mathbf{l})/2]=\sum_{\mathbf{q}} W_{12}(\mathbf{q})\Psi[(\mathbf{p}-\mathbf{l})/2-\mathbf{q}] \ . \tag{36}$$

The Heisenberg equations (11), (16), and (26), which have been derived from the e-h-γ Hamiltonian (3), transform to Eqs. (33)–(35) under the considered simplifications.

To understand the physical origin of the matrix element M_2, we rewrite the part of the Hamiltonian (29) which is responsible for the x-x attraction with Eq. (32):

$$\begin{aligned} H_{\text{int}} &= \tfrac{1}{2} \sum_{\sigma;\mathbf{p},\mathbf{l},\mathbf{q}} W_{\sigma,\sigma'}(\mathbf{q}) B^{\dagger}_{\sigma;\mathbf{p}} B^{\dagger}_{\sigma'\neq\sigma;\mathbf{l}} B_{\sigma'\neq\sigma;\mathbf{l}+\mathbf{q}} B_{\sigma;\mathbf{p}-\mathbf{q}} \\ &\simeq \tfrac{1}{4} \sum_{\sigma;\mathbf{p},\mathbf{l}} \frac{1}{\sqrt{V}} [M_2^*(\mathbf{p},\mathbf{l}) B^{\dagger}_{\sigma;\mathbf{p}} B^{\dagger}_{\sigma'\neq\sigma;\mathbf{l}} A_{\mathbf{p}+\mathbf{l}} + \text{H.c.}] \ . \end{aligned} \tag{37}$$

Thus the matrix element $M_2(\mathbf{p},\mathbf{k})$ characterizes the direct resonant coupling of the x's $\mathbf{p}$ and $\mathbf{k}$ of the initial polariton waves to form an m with $\mathbf{p}+\mathbf{k}$. This coupling is due to Coulomb interactions and can be resonant due to the polariton effect. In other words, the external electromagnetic waves with frequencies $\omega_1=\omega$ and $\omega_2=\omega_{\mathbf{k}}$ excite the polariton waves with finite x components, even if $\omega\leq\omega_t$ and $\omega_{\mathbf{k}}\leq\omega_t$ in order to allow a resonant m creation $\omega+\omega_{\mathbf{k}}\simeq\Omega^m_{\mathbf{p}+\mathbf{k}}$. Due to the polariton dispersion the energy can be conserved in the pairing of two x's of the polariton waves. Without the polariton effect the direct Coulombic x-x coupling to the m state is forbidden, because the total energy of two bare x's exceeds the m energy $\Omega^m_{\mathbf{p}+\mathbf{k}}$ by the corresponding m binding energy ϵ^m.

Thus the term $(1/\sqrt{V})M_2(\mathbf{p},\mathbf{k})A^{\dagger}_{\mathbf{p}+\mathbf{k}}B_{\mathbf{p}}B_{\mathbf{k}}$ rather than $(1/\sqrt{V})M_1(\mathbf{p},\mathbf{k})A^{\dagger}_{\mathbf{p}+\mathbf{k}}B_{\mathbf{p}}\alpha_{\mathbf{k}}$ describes the fundamental process of the optical creation of the m $\mathbf{p}+\mathbf{k}$. Equation (35) for the m operator reduces to the common Eq. (31) for the m-wave function $\Psi(\mathbf{l})$, if the last sum on the right-hand side of Eq. (35) is neglected.

In the phenomenological approach which uses the matrix element M_1,[19–23,26] γ, x, and m are treated as independent elementary bosons. The corresponding phenomenological Hamiltonian H_M has been derived intuitively, as stated in Ref. 27. In our model with the two different types of x's and γ's, this Hamiltonian would have the following form:

$$\begin{aligned} H_M = \sum_{\sigma;\mathbf{p}} \hbar \left[\omega^x_{\mathbf{p}} B^{\dagger}_{\sigma;\mathbf{p}} B_{\sigma;\mathbf{p}} + \omega^{\gamma}_{\mathbf{p}} \alpha^{\dagger}_{\sigma;\mathbf{p}} \alpha_{\sigma;\mathbf{p}} + \Omega^m_{\mathbf{p}} A^{\dagger}_{\mathbf{p}} A_{\mathbf{p}} + i\frac{\Omega_c}{2} (\alpha^{\dagger}_{\sigma;\mathbf{p}} B_{\sigma;\mathbf{p}} - B^{\dagger}_{\sigma;\mathbf{p}} \alpha_{\sigma;\mathbf{p}}) \right] \\ + \sum_{\sigma;\mathbf{p},\mathbf{l}} \left\{ \frac{1}{\sqrt{V}} M_1(\mathbf{p},\mathbf{l}) [A^{\dagger}_{\mathbf{p}+\mathbf{l}} \alpha_{\sigma;\mathbf{p}} B_{\sigma'\neq\sigma;\mathbf{l}} + \text{H.c.}] \right\} . \end{aligned} \tag{38}$$

Note, however, that the last term does not follow from the basic e-h-γ Hamiltonian as shown above. This three-particle interaction is instead constructed[1,28] with

$$\begin{aligned} \frac{1}{\sqrt{V}} M_1(\mathbf{p},\mathbf{l}) = \sum_{\sigma_1;\mathbf{q}} \Big\langle 0 \Big| A_{\mathbf{p}+\mathbf{l}} \left[-i\frac{\Omega_c}{2} B^{\dagger}_{\sigma_1;\mathbf{q}} \alpha_{\sigma_1;\mathbf{q}} \right] \\ \times B^{\dagger}_{\sigma'\neq\sigma;\mathbf{p}} \alpha^{\dagger}_{\sigma;\mathbf{l}} \Big| 0 \Big\rangle , \end{aligned} \tag{39}$$

where $|0\rangle$ denotes the crystal ground state. The proper convolution of pairs of operators on the right-hand side of Eq. (39) results in Eq. (1). The giant oscillator strength of the x-m optical conversion is now constructed in terms of the polariton transition between the m state $A^{\dagger}_{\mathbf{p}+\mathbf{k}}|0\rangle$ and the state with an x and a γ $B^{\dagger}_{\sigma;\mathbf{p}}\alpha^{\dagger}_{\sigma'\neq\sigma;\mathbf{k}}|0\rangle$. The closed set of the Heisenberg equations which follows from this phenomenological approach Eq. (38) with M_1 is given by

$$i\frac{\partial}{\partial t}\alpha_{\sigma;\mathbf{p}} = \omega^{\gamma}_{\mathbf{p}}\alpha_{\sigma;\mathbf{p}} + i\frac{\Omega_c}{2}B_{\sigma;\mathbf{p}} + \sum_{\mathbf{l}} \frac{1}{\sqrt{V}} M_1^*(\mathbf{p},\mathbf{l}) B^{\dagger}_{\sigma'\neq\sigma;\mathbf{l}} A_{\mathbf{p}+\mathbf{l}} , \tag{40}$$

$$i\frac{\partial}{\partial t}B_{\sigma;\mathbf{p}} = \omega^{x}_{\mathbf{p}}B_{\sigma;\mathbf{p}} - i\frac{\Omega_c}{2}\alpha_{\sigma;\mathbf{p}} + \sum_{\mathbf{l}} \frac{1}{\sqrt{V}} M_1^*(\mathbf{p},\mathbf{l}) \alpha^{\dagger}_{\sigma'\neq\sigma;\mathbf{l}} A_{\mathbf{p}+\mathbf{l}} , \tag{41}$$

$$\begin{aligned} i\frac{\partial}{\partial t}A_{\mathbf{K}} = \Omega^m_{\mathbf{K}}A_{\mathbf{K}} + \sum_{\sigma;\mathbf{l}} \frac{1}{\sqrt{V}} M_1(\mathbf{l}+\mathbf{K}/2,\mathbf{l}-\mathbf{K}/2) \\ \times \alpha_{\sigma;\mathbf{l}+\mathbf{K}/2} B_{\sigma'\neq\sigma;-\mathbf{l}+\mathbf{K}/2} \ . \end{aligned} \tag{42}$$

Both Eqs. (40) and (41), which describe the nonlinear propagation of the polariton wave $\mathbf{p}$, differ from the corresponding Eqs. (33) and (34) and are in contradiction to the e-h picture (3). The corresponding Eqs. (11) and (16) do not transform to Eqs. (40) and (41) under the considered assumptions. As will be shown below, Hamiltonians (29) and (38) lead to observable differences for, e.g., the x-m optical Stark effect and the 2γ m absorption. Only the m equations (35) and (42) have the identical form. In Sec. V, we will show that the description of the m luminescence is the same in both approaches.

The main problem with the phenomenological approach is due to the fact that x's and m's are not truly independent boson fields. According to representation (30), the degree of independence of these quasiparticles is given by the following commutation relations:

$$[A_{\mathbf{p}+\mathbf{k}}, B^{\dagger}_{\sigma;\mathbf{p}}] = \frac{1}{\sqrt{V}} \Psi \left[\frac{\mathbf{p}-\mathbf{k}}{2} \right] B_{\sigma' \neq \sigma;\mathbf{k}} \,,$$
$$[A_{\mathbf{K}}, A^{\dagger}_{\mathbf{P}}] = \delta_{\mathbf{K},\mathbf{P}} + \frac{1}{V} \sum_{l,\sigma} \Psi^{*}(\mathbf{l}) \Psi[(\mathbf{K}-\mathbf{P})/2 - \mathbf{l}] \times B^{\dagger}_{\sigma;-\mathbf{l}+\mathbf{P}/2} B_{\sigma;-\mathbf{l}+\mathbf{K}-\mathbf{P}/2} \,. \tag{43}$$

The first relation shows clearly the dependence between the m with momentum $\mathbf{p}+\mathbf{k}$ and the x's with $\mathbf{p}$ and $\mathbf{k}$. This lack of independence leads to the main objection against the use of the matrix element $M_1(\mathbf{p},\mathbf{k})$ for the description of the optical m creation.

III. LOW-INTENSITY EXCITON-BIEXCITON OPTICAL STARK EFFECT

The importance of the quantum dependence between the x's and the m's can be seen from the analysis of the low-intensity x-m optical Stark effect.[24,29] Here, one has to consider the dynamical properties of the γ-x-m system in the presence of a coherent polariton wave with $\mathbf{k}$ and $\omega_{\mathbf{k}}$ in the transparency region near the x absorption line. This intense wave will produce virtual electron-hole transitions, and therefore will drive the semiconductor to a nonequilibrium state. The pump wave with the circular polarization $\sigma=2$ is treated classically by replacing the x and γ operators of the given mode $\mathbf{k}$ by c numbers:

$$B_{2\mathbf{k}} \rightarrow \sqrt{V} P_{\mathbf{k}} \exp(-i\omega_{\mathbf{k}} t), \quad \alpha_{2\mathbf{k}} \rightarrow \sqrt{V} E_{\mathbf{k}} \exp(-i\omega_{\mathbf{k}} t) \,. \tag{44}$$

Here, $|P_{\mathbf{k}}|^2 \propto I_2$ and $|E_{\mathbf{k}}|^2 \propto I_2$ are the x and γ components of the polariton pump wave of the intensity I_2. The resulting nonequilibrium state makes it impossible to treat the x's and m's as being independent. Even in the thermodynamic limit $V \rightarrow \infty$ and $I_2=\text{const}$, finite corrections arise in the commutation relations (43). In our approach this difficulty is overcome by distinguishing mode $\mathbf{k}$ from other modes $\mathbf{p} \neq \mathbf{k}$ in the definition (30) of the m operator as well as in the initial Hamiltonian (29). This procedure corresponds to a Glauber transformation which removes the pump source from mode $\mathbf{k}$ by introducing a new vacuum state of the semiconductor in the presence of the polariton pump. Thus, for the m operator $A_{\mathbf{p}+\mathbf{k}}$ of Eq. (30), one has to use

$$A_{\mathbf{p}+\mathbf{k}} \rightarrow B_{1\mathbf{p}} P_{\mathbf{k}} \Psi[(\mathbf{p}-\mathbf{k})/2] e^{-i\omega_{\mathbf{k}} t} + \tilde{A}_{\mathbf{p}+\mathbf{k}} \,, \tag{45}$$

where the operator $\tilde{A}_{\mathbf{p}+\mathbf{k}}$ describes truly independent boson excitation. The Hamiltonian of Eq. (29) contains the following quadratic part of the operators of the $\sigma=1$ x's and γ's and m's:

$$H_1 = \sum_{\mathbf{p}} \hbar \left\{ \omega^{\gamma}_{\mathbf{p}} \alpha^{\dagger}_{1\mathbf{p}} \alpha_{1\mathbf{p}} + [\omega^{x}_{\mathbf{p}} + |P_{\mathbf{k}}|^2 V W_{12}(\mathbf{0})] B^{\dagger}_{1\mathbf{p}} B_{1\mathbf{p}} + \epsilon \tilde{A}^{\dagger}_{\mathbf{p}+\mathbf{k}} \tilde{A}_{\mathbf{p}+\mathbf{k}} + \left[i \frac{\Omega_c}{2} \alpha^{\dagger}_{1\mathbf{p}} B_{1\mathbf{p}} + \Delta[(\mathbf{p}-\mathbf{k})/2] P_{\mathbf{k}} e^{-i\omega_{\mathbf{k}} t} \tilde{A}^{\dagger}_{\mathbf{p}+\mathbf{k}} B_{1\mathbf{p}} + \text{H.c.} \right] \right\} , \tag{46}$$

where

$$\epsilon = \Omega^{m}_{\mathbf{K}} - \frac{1}{V} \sum_{l} (\omega^{x}_{-\mathbf{l}+\mathbf{K}/2} + \omega^{x}_{\mathbf{l}+\mathbf{K}/2}) |\Psi(\mathbf{l})|^2 \tag{47}$$

is the average potential energy of the x-x interaction in the m. We neglect all concentration-dependent corrections of the new vacuum state and consider only the resonant approximation of the m ground state. After a first canonical transformation with

$$S = \exp \left[i\omega_{\mathbf{k}} t \sum_{\sigma,\mathbf{p}} (\alpha^{\dagger}_{\sigma;\mathbf{p}} \alpha_{\sigma;\mathbf{p}} + B^{\dagger}_{\sigma;\mathbf{p}} B_{\sigma;\mathbf{p}}) \right] , \tag{48}$$

which removes the explicit time dependence of H_1 in Eq. (46), the Hamiltonian can be diagonalized with a second canonical transformation introducing new elementary excitations $\alpha'_{1;\mathbf{p}}$, $B'_{1;\mathbf{p}}$, and $A'_{\mathbf{p}+\mathbf{k}}$:

$$\begin{bmatrix} \alpha'_{1\mathbf{p}} \\ B'_{1\mathbf{p}} \\ A'_{\mathbf{p}+\mathbf{k}} \end{bmatrix} = (C_{ij}) \begin{bmatrix} \alpha_{1\mathbf{p}} \\ B_{1\mathbf{p}} \\ \tilde{A}_{\mathbf{p}+\mathbf{k}} \end{bmatrix} . \tag{49}$$

In this approximation, the transformation to the diagonal form treats both the x-γ and the x-x interactions exactly. The resulting frequency eigenvalues are

$$\nu^{\gamma} \tilde{\nu}^{x} \tilde{\nu}^{m} - |M_2(\mathbf{p},\mathbf{k}) P_{\mathbf{k}}|^2 \nu^{\gamma} - \tfrac{1}{2} \omega_t \Omega_c^2 \tilde{\nu}^{m} = 0 \,, \tag{50}$$

where

$$\nu^{\gamma} = \frac{c^2 p^2}{\epsilon_0} - \omega^2, \quad \tilde{\nu}^{x} = \omega^{x}_{\mathbf{p}} + \delta^{x}_{\mathbf{p}} - \omega \,,$$
$$\tilde{\nu}^{m} = \Omega^{m}_{\mathbf{p}+\mathbf{k}} + \delta^{m}_{\mathbf{p}+\mathbf{k}} - \omega - \omega_{\mathbf{k}} \,. \tag{51}$$

The nonresonant dynamical shifts $\delta^{x}_{\mathbf{p}}$ and $\delta^{m}_{\mathbf{p}+\mathbf{k}}$ of the x and m levels are determined by

$$\delta^{x}_{\mathbf{p}} = |P_{\mathbf{k}}|^2 V W_{12}(\mathbf{0}) \,,$$
$$\delta^{m}_{\mathbf{p}+\mathbf{k}} = |P_{\mathbf{k}}|^2 \left[V W_{12}(\mathbf{0}) + V W_{22}(\mathbf{0}) + \sum_{l} |\Psi(\mathbf{l})|^2 W_{22}[-\mathbf{l} + (\mathbf{p}-\mathbf{k})/2] \right] . \tag{52}$$

This dispersion Eq. (50) describes the splitting of the polariton and m frequencies caused by the polariton pump wave. The new dispersion is plotted in Fig. 1(b) and compared with the unperturbed polariton and m dispersions

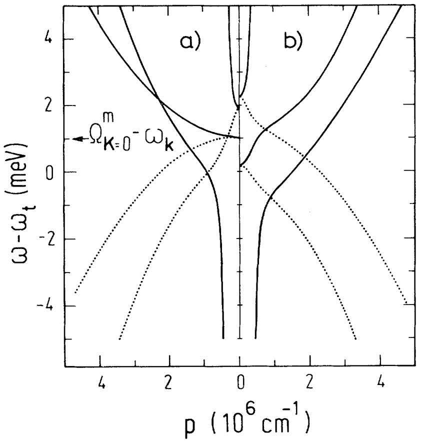

FIG. 1. Dispersion curves of the x-γ-m system of the semiconductor in the presence of the polariton pump wave (b) and unperturbed polariton and m dispersions (a). Real part of the wave number p (solid lines), imaginary part (dotted lines). The unperturbed m frequency $\Omega=\Omega_p$ is shifted by the pump frequency $\omega_{\mathbf{k}}$. The following CdS parameters have been used: $\omega_t=2.552$ eV, $\Omega^m_{\mathbf{K}=0}=5.100$ eV, $\omega_{lt}=1.9$ meV, $\epsilon_0=8.87$, $M=0.9m_0$, $|\Psi(0)|^2=2\times10^{-18}$ cm^3, and $\Delta(0)=-5$ meV. For the pump beam, $I_2=1$ MW/cm^2 and $\hbar\omega_{\mathbf{k}}=2.547$ meV have been used.

[Fig. 1(a)]. With the same procedure, one can derive the dispersion Eq. (50) directly from the Heisenberg Eqs. (33)–(35).

The weak polariton wave with $\mathbf{p}$, ω, and $\sigma=1$ acts as "probe wave" which tests the effective dynamical shift $\tilde{\delta}^x_{\mathbf{p}}$ of the x level, which is given by

$$\tilde{\delta}^x_{\mathbf{p}}=\delta^x_{\mathbf{p}}-\frac{|M_2(\mathbf{p},\mathbf{k})P_{\mathbf{k}}|^2}{\Omega^m_{\mathbf{p}+\mathbf{k}}+\delta^m_{\mathbf{p}+\mathbf{k}}-\omega_{\mathbf{k}}-\omega^x_{\mathbf{p}}} \, . \tag{53}$$

The dynamical shift of the x level has two contributions. The first, $\delta^x_{\mathbf{p}}$ [Eq. (52)], is the shift due to the renormalization of the vacuum state by the pump wave. It stems from nonresonant Coulombic attraction of the probe x with $\mathbf{p},\sigma=1$ and the x with $\mathbf{k},\sigma=2$ of the pump wave. The more important second term in Eq. (53) for the low-intensity x-m optical Stark effect is due to the resonant x-x interaction resulting in virtual m formation with momentum $\mathbf{p}+\mathbf{k}$ from probe $\mathbf{p},\sigma=1$ and pump $\mathbf{k},\sigma=2$ x's. The efficiency of this process is determined by the matrix element $M_2(\mathbf{p},\mathbf{k})$. As a result, the new length parameter a_m appears naturally. The second term in Eq. (53) dominates because of the resonant denominator and results in a redshift of the x level for the most interesting frequency range $\omega_{\mathbf{k}}+\omega^x_{\mathbf{p}}<\Omega^m_{\mathbf{p}+\mathbf{k}}$.

First, theoretical studies of the dynamical x-γ-m spectra renormalization within the phenomenological approach have been given in Refs. 23 and 24. Subsequent similar results have been obtained in connection with the dynamical x-m Stark effect.[27] The corresponding dispersion equation can be obtained from Eqs. (40)–(42) or from the treatment of the phenomenological Hamiltonian (38):

$$\nu^{\text{phot}}\nu^{\text{ex}}\nu^m-2\omega_t|M_1(\mathbf{p},\mathbf{k})P_{\mathbf{k}}|^2\nu^{\text{ex}} -|M_1(\mathbf{p},\mathbf{k})E_{\mathbf{k}}|^2\nu^{\text{phot}}-\tfrac{1}{2}\omega_t\Omega_c^2\nu^m=0 \, , \tag{54}$$

where

$$\nu^{\text{ex}}=\omega^x_{\mathbf{p}}-\omega, \quad \nu^m=\Omega^m_{\mathbf{p}+\mathbf{k}}-\omega-\omega_{\mathbf{k}} \, . \tag{55}$$

In this case, the x-m optical Stark effect is determined by the matrix element $M_1(\mathbf{p},\mathbf{k})$. The same definition of the strength of the x-m optical conversion in terms of M_1 has been used in the analysis of the optical Stark effect in Refs. 30 and 31.

The derivation of Eq. (54) from the phenomenological Hamiltonian (38) is inconsistent, because the c-number substitution (44) is used only for the x and γ operators, but not for the m operators which are considered to be independent. The same objection against the use of the matrix element $M_1(\mathbf{p},\mathbf{k})$ can be obtained directly from its definition in Eq. (39). For a coherent polaritons pump wave, the m state $A^\dagger_{\mathbf{p}+\mathbf{k}}|0\rangle$ and the x-γ state $B^\dagger_{1\mathbf{p}}\alpha^\dagger_{2\mathbf{k}}|0\rangle$ are not independent, orthogonal states, as can be seen from the definition of $A_{\mathbf{K}}$ in Eq. (30). We conclude that the matrix element $M_1(\mathbf{p},\mathbf{k})$ of Eq. (39) for transitions between two dependent quantum states does not describe a real m creation process correctly.

For the derivation of both dispersions, Eqs. (50) and (54), the nonresonant part of the x-γ interaction has been taken into account in the corresponding γ equations. In other words, instead of Eqs. (33) and (40) the corresponding Maxwell equations have been used. This procedure ensures the correct limit for all dispersion branches $\omega=\omega_i(p)$ for $p\rightarrow0$.

In conclusion, we state that the described self-consistent theory of the x-m optical Stark effect considers x's without internal structure. However, the main results will be preserved, even if one uses the exact e-h picture instead of the x approximation. This theory has been developed in Ref. 24. In this theory both x's and m's have been treated as two- and four-particle e-h complexes in accordance with Eqs. (8) and (9). The new vacuum state and the m's are introduced self-consistently in the presence of a coherent polariton pump wave. The resulting dispersion Eq. (50) describes the x-m optical Stark. The crucial matrix element $M_2(\mathbf{p},\mathbf{k})$ has been expressed explicitly in terms of the x- and m-wave functions $\phi(\mathbf{l})$ and $\Gamma_J(\mathbf{p},\mathbf{l},\mathbf{q})$, respectively. For a weakly bound m, M_2 is given by the simple form of Eq. (36).

IV. TWO-PHOTON BIEXCITON ABSORPTION

The 2γ absorption due to m excitation can be calculated directly from the derived dispersion, i.e., Eq. (50) for the self-consistent theory, or Eq. (54) for the phenomenological one. In order to get a more realistic description we introduce the inverse x and m lifetimes γ^x and γ^m by substituting in these dispersion relations: $\omega^x_{\mathbf{p}}\rightarrow\omega^x_{\mathbf{p}}-i\gamma^x$ and $\Omega^m_{\mathbf{p}+\mathbf{k}}\rightarrow\Omega^m_{\mathbf{p}+\mathbf{k}}-i\gamma^m$. Now a natural threshold for the existence of the x-m optical Stark effect arises. For $|\omega-\omega_t|\geq\omega_{lt}$, this threshold is determined by the condition

$$\frac{\omega_{lt}|M_2(\mathbf{p},\mathbf{k})P_{\mathbf{k}}|^2}{(\omega-\omega_t)^2} \geq \gamma^m,\gamma^x \ . \tag{56}$$

Below this threshold for the pump x amplitude and the corresponding pump intensity $I_{2,c}$, the dissipative rather than dispersive part of the corresponding nonlinear process dominates. Thus in this low-intensity limit one can study the 2γ m absorption.

The absorption coefficient of the probe beam with $\sigma=1$ and $\mathbf{p},\omega \simeq \Omega^m_{\mathbf{p}+\mathbf{k}}-\omega_{\mathbf{k}}$ is given by twice the imaginary part of the wave number p, i.e., $\alpha(\omega)=2\,\mathrm{Im}p(\omega,\omega_{\mathbf{k}},I_2,\gamma^x,\gamma^m)$. Thus $\alpha(\omega)$ can be obtained from Eqs. (50) or (54) (see also Fig. 1) for a sufficiently small pump wave intensity $I_2 \rightarrow 0$, to be below the threshold of the x-m optical Stark effect. More accurately, one has to define the pure m absorption coefficient $K^{(2)}_m(\omega,\omega_{\mathbf{k}})$ by subtracting the usual linear x absorption:

$$\begin{aligned} K^{(2)}_m(\omega,\omega_{\mathbf{k}}) &= 2\,\mathrm{Im}p(\omega,\omega_{\mathbf{k}},I_2<I_{2,c},\gamma^x,\gamma^m) \\ &\quad -2\,\mathrm{Im}p(\omega,\omega_{\mathbf{k}},I_2<I_{2,c},\gamma^x,\gamma^m \rightarrow 0) \ . \end{aligned} \tag{57}$$

Already, within the framework of the initial Heisenberg Eqs. (33)–(35) or (40)–(42), one sees the different mechanisms for the two-γ absorption in the self-consistent and phenomenological model, respectively. According to Eqs. (33) and (34) the nonlinear polariton absorption is governed by the third term on the right-hand side of Eq. (34). This term is responsible for the x-x interaction. In turn, Eqs. (40) and (41) describe the 2γ absorption in the phenomenological theory due to the matrix element $M_1(\mathbf{p},\mathbf{l})$. The appearance of the nonlinear term in Eq. (40) for γ is unphysical. The proper nonlinear term responsible for the 2γ absorption should appear only in the x equation. This statement follows from the fact that in the considered resonant m optical nonlinearity only the interaction between polarizations is important and formally can be proved easily from the comparison of Eq. (40) with Eq. (11), which follows directly from the basic e-h-γ Hamiltonian of Eq. (3).

Instead of absorption coefficient α, we define the coefficient $K^{(2)}_m(\omega,\omega_{\mathbf{k}})$ of the m 2γ absorption as usual by

$$\frac{d}{dz}I_\sigma = -K^{(2)}_m(\omega,\omega_{\mathbf{k}})\sqrt{I_1}\sqrt{I_2} \ , \tag{58}$$

where I_σ are the intensities of the two circularly polarized light beams, with $\omega_{\sigma=1}=\omega$ and $\omega_{\sigma=2}=\omega_{\mathbf{k}}$, which penetrate the crystal through a surface and which are absorbed under the condition $\omega+\omega_{\mathbf{k}} \simeq \Omega^m_{\mathbf{p}+\mathbf{k}}$. From Eq. (50), the self-consistent theory yields, for the 2γ absorption coefficient,

$$K^{(2)}_m(\omega,\omega_{\mathbf{k}}) = \frac{\epsilon_0}{c^2}\frac{|\Delta[(\mathbf{p}-\mathbf{k})/2]|^2\Omega_c^4}{8\hbar(\omega_{\mathbf{k}}-\omega_t)^2(\omega-\omega_t)^2}\left[\frac{I_1}{\omega}\right]^{1/2}\left[\frac{I_2}{\omega_{\mathbf{k}}}\right]^{1/2}\frac{\gamma^m}{(\Omega^m_{\mathbf{p}+\mathbf{k}}-\omega_{\mathbf{k}}-\omega)^2+(\gamma^m)^2} \ , \tag{59}$$

while the dispersion of the phenomenological theory Eq. (54) yields

$$\begin{aligned} \widetilde{K}^{(2)}_m(\omega,\omega_{\mathbf{k}}) &= \frac{\epsilon_0}{c^2}\frac{|\Psi[(\mathbf{p}-\mathbf{k})/2]|^2\Omega_c^4}{8\hbar(\omega_{\mathbf{k}}-\omega_t)^2(\omega-\omega_t)^2} \\ &\quad \times[(\omega_{\mathbf{k}}-\omega_t)^2+(\omega-\omega_t)^2] \\ &\quad \times\left[\frac{I_1}{\omega}\right]^{1/2}\left[\frac{I_2}{\omega_{\mathbf{k}}}\right]^{1/2} \\ &\quad \times\frac{\gamma^m}{(\Omega^m_{\mathbf{p}+\mathbf{k}}-\omega_{\mathbf{k}}-\omega)^2+(\gamma^m)^2} \ . \end{aligned} \tag{60}$$

Both spectra $K^{(2)}_m$ [Eq. (59)] and $\widetilde{K}^{(2)}_m$ [Eq. (60)] have been derived for the conditions $\Omega_{\mathrm{res}} \geq |\omega-\omega_t| \geq \omega_{lt}$ and $\Omega_{\mathrm{res}} \geq |\omega_{\mathbf{k}}-\omega_t| \geq \omega_{lt}$, where the parameter Ω_{res} determines the boundary of the frequency range for which a single intermediate resonant x level ($n=1$) for 2γ m absorption can be considered. Small corrections which are proportional to

$$\eta_{i=1,2} = \frac{2\gamma^x}{\gamma^m}\frac{|\Omega^m_{\mathbf{K}=0}-\omega-\omega_{\mathbf{k}}|}{|\omega_t-\omega_{i=1,2}|} \ll 1 \ , \tag{61}$$

have been neglected in both results, i.e., in Eqs. (59) and (60).

In the derivation of the 2γ absorption coefficients $K^{(2)}_m(\omega,\omega_{\mathbf{k}})$ and $\widetilde{K}^{(2)}_m(\omega,\omega_{\mathbf{k}})$, the polariton character of the propagation of the both electromagnetic waves has been explicitly taken into account, in contrast to the phenomenological approach.[1] The main corrections arising from the polariton effect are connected with the composite x-γ structure of a polariton wave, as well as with large difference between the polariton group velocity and the "background velocity" $c/\sqrt{\epsilon_0}$. As already stated above, both of these corrections are important in the frequency intervals $|\omega_{\mathbf{k}}-\omega_t| \leq \Omega_c$ and $|\omega-\omega_t| \leq \Omega_c$.

In order to identify the origin of the main differences between the two results $K^{(2)}_m$ and $\widetilde{K}^{(2)}_m$, we return to the analysis of the basic elementary processes of the 2γ absorption due to m formation. The schematic representations of two different models of this process are given in Figs. 2 and 3. In these graphs, the solid and wavy lines refer to an x and γ propagation, respectively, the bold dot symbolizes an x-γ (polariton) transition, whereas the box Γ represents the m state. According to the traditional phenomenological concept (see Fig. 2), a 2γ m absorption $\widetilde{K}^{(2)}_m$ is determined by the matrix element $M_1(\mathbf{p},\mathbf{k})$ of Eq. (39). As already discussed, one immediately finds Eq. (1) for the matrix element $M_1(\mathbf{p},\mathbf{k})$ from the convolution of the pairs of operators on the right-hand side of Eq. (39). This procedure implies that the two x's in the created m have the same wave vectors $\mathbf{p}$ and $\mathbf{k}$ as initial γ's. According to the well-known Hopfield concept, this momentum conservation does not hold for true absorption. For true absorption (and not only virtual excitation), a real scattering process has to occur. This scattering breaks the quantum coherence and makes the excitation process irreversible. In our approach the Coulombic scattering of the two x's which form the m is in combination with the polariton dispersion responsible for the real 2γ absorption (see Fig. 3).

The conservation of the initial momenta $\mathbf{p}$ and $\mathbf{k}$ of the two x's in the phenomenological picture is a manifestation of the nonorthogonality of the states $A^{\dagger}_{\mathbf{p}+\mathbf{k}}|0\rangle$ and $B^{\dagger}_{1,2;\mathbf{p}}\alpha^{\dagger}_{2,1;\mathbf{k}}|0\rangle$. In the self-consistent approach the interior structure of the m is treated explicitly. The scattering process which is responsible for a 2γ absorption $K^{(2)}_m$ is introduced by taking into account the whole set of the ladder diagrams (see Fig. 3). Here, the dashed lines refer to the Coulombic potential $W_{12}(\mathbf{q})$. According to Eq. (59), one obtains the true m pole in the corresponding scattering amplitude of the x's.

The 2γ absorption coefficients $K^{(2)}_m(\omega,\omega_{\mathbf{k}})$ and $\tilde{K}^{(2)}_m(\omega,\omega_{\mathbf{k}})$ both have the proper symmetry with respect to the frequencies ω and $\omega_{\mathbf{k}}$. The frequency band of the 2γ absorption due to m formation represents the usual Lorentz resonance denominator with the central frequency $\Omega=\Omega^m_{\mathbf{p}+\mathbf{k}}=\omega+\omega_{\mathbf{k}}$. The frequency width is determined by the m scattering parameter γ^m. This scattering parameter γ^m in our model is given by the inverse m radiative lifetime. In Sec. V, it is calculated from first principles with the Hamiltonian of Eq. (29).

From the definition of the matrix element Δ and the m Schrödinger equation [Eq. (31)], one obtains

$$\Delta[(\mathbf{p}-\mathbf{k})/2]=[-\epsilon^m-\hbar^2(\mathbf{p}-\mathbf{k})^2/4M]\Psi[(\mathbf{p}-\mathbf{k})/2] \ . \tag{62}$$

Because the polariton wave numbers $\mathbf{p}$ and $\mathbf{k}$ refer to 2γ absorption to the optical range, the second term in the square brackets of Eq. (62) can be neglected in the first approximation. The main difference between the self-consistent and the phenomenological 2γ absorption spectra of Eqs. (59) and (60), respectively, is their frequency dependence via the factors $(\omega_{\mathbf{k}}-\omega_t)^2$ and $(\omega-\omega_t)^2$. The ratio $R(\omega,\omega_{\mathbf{k}})$ of two spectra has the simple form

$$R(\omega,\omega_{\mathbf{k}})=\frac{\tilde{K}^{(2)}_m(\omega,\omega_{\mathbf{k}})}{K^{(2)}_m(\omega,\omega_{\mathbf{k}})}=\frac{(\omega_{\mathbf{k}}-\omega_t)^2+(\omega-\omega_t)^2}{(\epsilon^m)^2} \ . \tag{63}$$

The phenomenological 2γ absorption $\tilde{K}^{(2)}_m(\omega,\omega_{\mathbf{k}})$ is determined by the process

$$\gamma+\gamma\rightarrow x+\gamma\rightarrow m \ ,$$

while the self-consistent 2γ absorption $K^{(2)}_m(\omega,\omega_{\mathbf{k}})$ follows the scheme

$$\gamma+\gamma\rightarrow x+x\rightarrow m \ .$$

Of course, in a polariton representation, both approaches are described as

$$\gamma+\gamma\rightarrow \text{polariton}+\text{polariton}\rightarrow m \ .$$

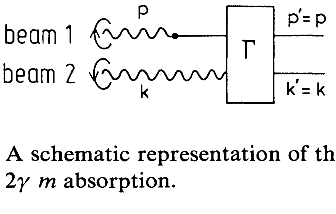

FIG. 2. A schematic representation of the phenomenological model of a 2γ m absorption.

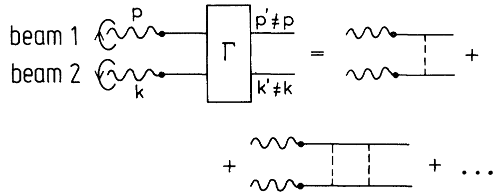

FIG. 3. A schematic representation of the self-consistent model of a 2γ m absorption.

However, the efficiencies of the 2γ absorption, i.e, the corresponding matrix elements of the conversion of the two polaritons to an m, are different for the two approaches.

The role of real scattering processes in the Hopfield concept of true absorption is a more delicate question for the 2γ absorption of the m than for the linear absorption of the x. In the latter case the scattering system, i.e., impurities or phonons, represents an "external system" for the scattered polariton $\mathbf{k}$. If, after the first optical transition γ $\mathbf{p}\rightarrow x$ $\mathbf{p}$, the created x $\mathbf{p}$ is scattered before the inverse optical transition x $\mathbf{p}\rightarrow\gamma$ $\mathbf{p}$ takes place, the usual semiclassical approach to the x absorption is valid. In such a case the integrated x absorption coefficient $\int d\omega\,\alpha_x(\omega)$ is proportional to Ω_c^2, i.e., is determined only by the strength of the x-γ optical transition. By changing the temperature or the impurity concentration, one can move from this regime to the polariton regime in which even the integrated x absorption coefficient depends both on the x-γ matrix element Ω_c and on the x scattering parameter γ^x.

For a 2γ absorption of the m, the situation is different, because the created m $\mathbf{p}+\mathbf{k}$ itself represents an "internal scattering system" for the initial polaritons $\mathbf{p}$ and $\mathbf{k}$. So the question about the balance between a polariton effect and an interior scattering process arises naturally, as both processes are resonant. According to the phenomenological approach the two constituent x's $\mathbf{p}$ and $\mathbf{k}$ of the optically created m $\mathbf{p}+\mathbf{k}$ automatically scatter each other before the inverse optical transition x $\mathbf{p}(\mathbf{k})\rightarrow\gamma$ $\mathbf{p}(\mathbf{k})$ occurs. The relation of Eq. (63) shows that this statement is correct only if one of the frequencies ω or $\omega_{\mathbf{k}}$ approaches the x energy ω_t (e.g., if $\omega-\omega_t\rightarrow 0$ and $\omega_t-\omega_{\mathbf{k}}\rightarrow\epsilon^m$). In this case $R(\omega,\omega_{\mathbf{k}})\rightarrow 1$, so that both results $K^{(2)}_m(\omega,\omega_{\mathbf{k}})$ and $\tilde{K}^{(2)}_m(\omega,\omega_{\mathbf{k}})$ yield the same limit in spite of the fact that they have been derived for the frequency range $|\omega-\omega_t|\geq\omega_{lt}, |\omega_{\mathbf{k}}-\omega_t|\geq\omega_{lt}$. The polariton wave $\mathbf{p}$ with the frequency $\omega\rightarrow\omega_t$ represents a pure x wave with a small "mechanical" group velocity. One can compare the x's of this wave with impurities and the m state with an impurity-bound x state. In this limit $(\omega-\omega_t\rightarrow 0)$, both approaches give the same result in agreement with the Rashba theory (see, e.g., Ref. 32) of the giant oscillator strength of the shallow impurity-bound states. Using methods of the scattering theory, Hopfield[33] showed that an x $\mathbf{p}$ resonantly bound to an impurity always has to be scattered before the reemission of a γ with the same momentum $\mathbf{p}$ occurs.

The different dependencies of the absorption coefficients $K_m^{(2)}(\omega,\omega_\mathbf{k})$ and $\tilde{K}_m^{(2)}(\omega,\omega_\mathbf{k})$ on the frequencies ω and $\omega_\mathbf{k}$ can be distinguished experimentally. CuBr and CuCl semiconductors are best suited for such studies. The calculated spectra of the integrated 2γ absorption coefficients $K_{\text{int}}(\omega)=\int d\omega_\mathbf{k} K_m^{(2)}(\omega,\omega_\mathbf{k})$ and $\tilde{K}_{\text{int}}(\omega)$ are shown in Fig. 4. The two curves [solid line, $K_{\text{int}}(\omega)$; dashed line, $\tilde{K}_{\text{int}}(\omega)$] deviate most strongly in the degeneracy region $\omega=\omega_\mathbf{k}=\frac{1}{2}\Omega_{\mathbf{K}=0}^m$ and for the absorption in the spectral wing $\omega<\omega_t-\epsilon^m(\omega_\mathbf{k}>\omega_t)$.

The degenerate absorption corresponds to a minimum of the both curves (points O and O', respectively) at the range $0\le\omega_t-\omega\le\epsilon^m$. According to Eq. (59), one can obtain the following approximation in the spectral vicinity of the degenerate absorption:

$$K_{\text{int}}(\omega)=K_{\text{int}}^{(0)}[1+8(\Delta/\epsilon^m)^2] , \tag{64}$$

whereas the phenomenological approach of Eq. (60) yields

$$\tilde{K}_{\text{int}}(\omega)=\tilde{K}_{\text{int}}^{(0)}[1+12(\Delta\omega/\epsilon^m)^2] . \tag{65}$$

Here, $K_{\text{int}}^{(0)}$ and $\tilde{K}_{\text{int}}^{(0)}$ are the integrated absorption coefficients for the degenerate case, $\Delta\omega=\frac{1}{2}\Omega_{\mathbf{K}=0}^m-\omega$. If one knows the m binding energy sufficiently well, the differences between Eqs. (64) and (65) can be tested experimentally. In the low-frequency tail $\omega<\omega_t-\epsilon^m$, Eq. (60) strongly overestimates the m absorption. Interesting preliminary experimental investigations of CuCl by Nagasawa and Hasuo[34] do not yet allow for a final decision between the self-consistent and the phenomenological 2γ absorption spectrum of m.

Another important difference between $K_m^{(2)}(\omega,\omega_\mathbf{k})$ and $\tilde{K}_m^{(2)}(\omega,\omega_\mathbf{k})$ is connected with their dependencies on the 2γ absorption geometry, i.e., on the angle θ between vectors $\mathbf{p}$ and $\mathbf{k}$. Although such a dependence appears only as a small correction to $K_m^{(2)}(\omega,\omega_\mathbf{k})$ it is in principle different in the two spectra. For example, one can consider the potential $W_{12}(\mathbf{l}-\mathbf{l}')$ of the x-x attraction as a Coulomb potential and approximate the m ground-state wave function by $\Psi(l)=8\sqrt{\pi a_m^3}/(1+a_m^2 l^2)^2$. With this approximation one finds, for the degenerate case,

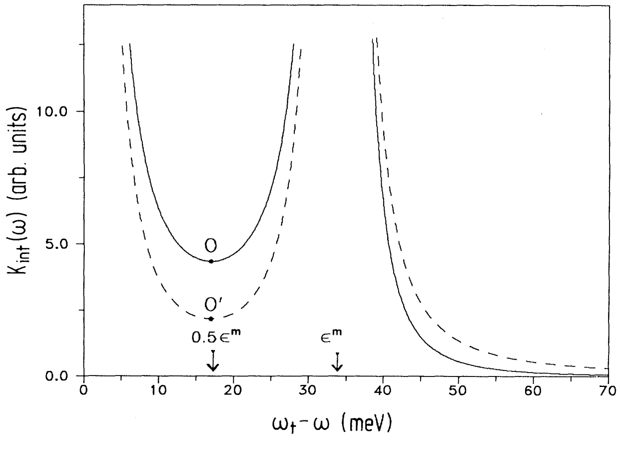

FIG. 4. The frequency dependence of the integral 2γ m absorption coefficient $\mathbf{K}_{\text{int}}(\omega)=\int d\omega_\mathbf{k} K_m^{(2)}(\omega,\omega_\mathbf{k})$. Self-consistent model Eq. (59) (solid line), phenomenological model Eq. (60) (dashed line) for the CuCl parameter $\omega_t=3.2022$ eV, $\omega_{lt}=5.7$ meV, and $\epsilon^m=34$ meV.

$$K_m^{(2)}(\omega,\omega)=K_\parallel^{(2)}(1-6a_m^2k^2\sin^2\theta/2) \tag{66}$$

and

$$\tilde{K}_m^{(2)}(\omega,\omega)=\tilde{K}_\parallel^{(2)}(1-2a_m^2k^2\sin^2\theta/2) . \tag{67}$$

Here, $K_\parallel^{(2)}$ and $\tilde{K}_\parallel^{(2)}$ are the m absorption coefficients for two copropagating polaritons, i.e., when $\mathbf{p}=\mathbf{k}$. The angle dependence of $K_m^{(2)}(\omega,\omega)$ reaches about 1% for CuCl and about 10% for CdS crystal.

For real experiments, corrections for the intensities I_σ of the electromagnetic waves inside the crystal are necessary. For normal incidence of both waves on the surface of the crystal, the external intensities $I_{0,\sigma}$ are connected with intensities I_σ by the relation

$$I_\sigma=\frac{4n_\sigma}{(n_\sigma+1)^2}I_{0,\sigma} , \tag{68}$$

where the corresponding polariton refractive index n_σ is given by

$$n_\sigma=\sqrt{\epsilon_0[1+\omega_{lt}/(\omega_t-\omega_\sigma)]}\simeq\sqrt{\epsilon_0} , \tag{69}$$

and $\omega_{\sigma=1,2}=\omega,\omega_\mathbf{k}$.

To summarize, the phenomenological theory is inaccurate because it neglects an important quantum dependence of x's and m's. This failure leads to the incorrect introduction of the matrix element $M_1(\mathbf{p},\mathbf{k})$ and the corresponding picture of the optical creation of m. The phenomenological approach yields the correct results for the 2γ absorption of m and the x-m optical Stark effect only if one of the frequencies ω or $\omega_\mathbf{k}$ of the polariton waves tends to the x level position ω_t. Concerning the general characteristic of the matrix element $M_2(\mathbf{p},\mathbf{k})$ and the corresponding scenario for the optical excitation of an m, another comment may be helpful. In the self-consistent approach this matrix element also characterizes the giant oscillator strength of the optical conversion of the two polaritons to an m in the usual sense.[1,28,32] Our results deal with the *efficiency* of the nonlinear optical processes. All results concerning the *spectral characteristics,* i.e., the spectral positions of the signals in hyper-Raman scattering via m's, the spectral position of the 2γ absorption m peak, etc., evidently are the same in both formulations.

V. LUMINESCENCE OF BIEXCITONS

One of the main conclusions of the general scattering theory is the close formal relation between the total resonant cross section and the lifetime of the corresponding bound metastable state.[35] For our problems, the resonant x-x scattering causes the 2γ m absorption. Thus the 2γ m absorption and the inverse process, the m luminescence, are different manifestations of the same basic scattering process. In the framework of our dynamical model, the parameter γ^m in Eqs. (59) and (60) for the 2γ m absorption corresponds to the inverse m *radiative* lifetime. In order to make the description of the 2γ absorption completely self-consistent, one has to find this inverse radiative lifetime $\gamma^m_{\mathbf{p}+\mathbf{k}}$ of the m $\mathbf{p}+\mathbf{k}$ through the parameters of the initial Hamiltonian (29).

The radiative lifetime of an m state can be determined directly within the dynamic Eqs. (33)–(35). Here, the third nonlinear term on the right-hand side of Eq. (34) can be neglected. Expressing the γ operators $\alpha_{\sigma;\mathbf{p}}$ in terms of the x operators $B_{\sigma;\mathbf{p}}$ by means of Eqs. (33) and (34), and substituting these expressions in the right-hand side of Eq. (35), one obtains a Schrödinger equation for the m-wave function $\tilde{\Psi}(l)$ which includes polariton effects. Diagram techniques allow a still shorter derivation of this equation. Both sets of Eqs. (33)–(35) and (40)–(42) lead to an identical description of the luminescence process.

Following Ref. 36, one studies the poles of the vertex function which describes the x-x four-particle interaction for the description of an m bound state. The corresponding homogeneous Bethe-Salpeter equation for the vertex function $\Gamma(l,l'';r)$ can be written in the form

$$\Gamma(l,l'';K)=\sum_{l'} W_{12}(l-l')\tilde{G}(l',K/2)\Gamma(l',l'';K) \ . \tag{70}$$

Here, $l=\{\mathbf{l},\omega\}$ and $l''=\{\mathbf{l}'',\omega''\}$ are the reduced relative momenta and frequencies of two x's before and after the interaction act. $K=\{\mathbf{K},\Omega\}$ describes the conserved total momentum and energy. The $2x$ Green's function $\tilde{G}(l',K/2)$ is given by the convolution

$$\tilde{G}(l',K/2)=-\frac{i}{2\pi}\int d\omega' G(-l'+K/2)G(l'+K/2) \ , \tag{71}$$

where $G(\pm l'+K/2)$is the x Green's function. With the property of the vertex function

$$\tilde{G}(l,K/2)\tilde{G}(l'',-K/2)\Gamma(l,l'';K)\propto\tilde{\Psi}(l)\tilde{\Psi}(l'') \ , \tag{72}$$

one gets, for $K=(\mathbf{K}=0,\Omega=\tilde{\Omega}^m_{\mathbf{K}=0})$,

$$\tilde{G}^{-1}(\mathbf{p},\tfrac{1}{2}\tilde{\Omega}^m_{\mathbf{K}=0})\tilde{\Psi}(\mathbf{p})-\sum_{\mathbf{p}'} W_{12}(\mathbf{p}-\mathbf{p}')\tilde{\Psi}(\mathbf{p}')=0 \ . \tag{73}$$

Equation (73) is the m-wave equation which describes the relative motion in the bound state of two x's with momenta $\mathbf{p}$ and $\mathbf{k}=-\mathbf{p}$.

In order to evaluate Eq. (71), we express the x Green's function $G(l)$ in terms of the x and γ free-particle Green's functions

$$G_0(\mathbf{p},\omega)=\frac{1}{\omega-\omega^x_{\mathbf{p}}+i\delta}, \quad D_0(\mathbf{p},\omega)=\frac{1}{\omega-\omega^\gamma_{\mathbf{p}}+i\delta} \tag{74}$$

by the relation

$$\begin{aligned} G(\mathbf{p},\omega)&=\frac{D_0^{-1}(\mathbf{p},\omega)}{G_0^{-1}(\mathbf{p},\omega)D_0^{-1}(\mathbf{p},\omega)-\frac{1}{4}\Omega_c^2} \\ &=\frac{\phi^+(\mathbf{p},\omega)}{\omega-\omega^+_{\mathbf{p}}+i\delta}+\frac{\phi^-(\mathbf{p},\omega)}{\omega-\omega^-_{\mathbf{p}}+i\delta} \ , \end{aligned} \tag{75}$$

where

$$\phi^{+(-)}(\mathbf{p},\omega)=\frac{\omega^{+(-)}_{\mathbf{p}}(\omega-\omega^\gamma_{\mathbf{p}})}{\omega(\omega^{+(-)}_{\mathbf{p}}-\omega^{-(+)}_{\mathbf{p}})} \ . \tag{76}$$

Here, $\omega^\pm_{\mathbf{p}}$ are the frequencies of the upper $(+)$ and the lower $(-)$ polariton branches, respectively, i.e., the roots of the polariton dispersion

$$\frac{c^2p^2}{\epsilon_0\omega^2}=1+\frac{\Omega_c^2}{\omega_t^2+\hbar\omega_t p^2/M-\omega^2} \ . \tag{77}$$

In the final expression we include the nonresonant part of the x-γ interaction. The functions $\phi^{+(-)}(\mathbf{p},\omega)$ satisfy to the following conditions:

$$\begin{aligned} &\phi^+(\mathbf{p},\omega^+_{\mathbf{p}})\geq 0, \quad \phi^-(\mathbf{p},\omega^-_{\mathbf{p}})\geq 0 \ , \\ &\phi^+(\mathbf{p},\omega^+_{\mathbf{p}})+\phi^-(\mathbf{p},\omega^-_{\mathbf{p}})=1 \ . \end{aligned} \tag{78}$$

These functions characterize the distribution of the x component between the two polariton dispersion branches. Thus Eq. (78) is the "sum rule" for the x.

With the x-polariton Green's function (75), one finds

$$\begin{aligned} \tilde{G}(\mathbf{p},\tfrac{1}{2}\tilde{\Omega}^m_{\mathbf{K}=0})&=-\frac{i}{2\pi}\int d\omega' G(-\mathbf{p},-\omega'+\tfrac{1}{2}\tilde{\Omega}^m_{\mathbf{K}=0})G(\mathbf{p},\omega'+\tfrac{1}{2}\tilde{\Omega}^m_{\mathbf{K}=0}) \\ &=\left[\frac{\phi^+(\mathbf{p},\omega^+_{\mathbf{p}})\phi^+(-\mathbf{p},\tilde{\Omega}^m_{\mathbf{K}=0}-\omega^+_{\mathbf{p}})}{\tilde{\Omega}^m_{\mathbf{K}=0}-\omega^+_{\mathbf{p}}-\omega^+_{-\mathbf{p}}+i\delta}+\frac{\phi^+(\mathbf{p},\omega^+_{\mathbf{p}})\phi^-(-\mathbf{p},\tilde{\Omega}^m_{\mathbf{K}=0}-\omega^+_{\mathbf{p}})}{\tilde{\Omega}^m_{\mathbf{K}=0}-\omega^+_{\mathbf{p}}-\omega^-_{-\mathbf{p}}+i\delta}\right. \\ &\quad\left.+\frac{\phi^-(\mathbf{p},\omega^-_{\mathbf{p}})\phi^+(-\mathbf{p},\tilde{\Omega}^m_{\mathbf{K}=0}-\omega^-_{\mathbf{p}})}{\tilde{\Omega}^m_{\mathbf{K}=0}-\omega^-_{\mathbf{p}}-\omega^+_{-\mathbf{p}}+i\delta}+\frac{\phi^-(\mathbf{p},\omega^-_{\mathbf{p}})\phi^-(-\mathbf{p},\tilde{\Omega}^m_{\mathbf{K}=0}-\omega^-_{\mathbf{p}})}{\tilde{\Omega}^m_{\mathbf{K}=0}-\omega^-_{\mathbf{p}}-\omega^-_{-\mathbf{p}}+i\delta}\right] \ . \end{aligned} \tag{79}$$

Thus Eq. (73) with function $\tilde{G}(\mathbf{p},\frac{1}{2}\tilde{\Omega}^m_{\mathbf{K}=0})$ given by Eq. (79) is the final general form of an m Schrödinger equation which describes m with the translation momentum $\mathbf{K}=0$. The generalization of this result for an arbitrary total momentum $\mathbf{K}$ is obtained by replacing in Eqs. (73) and (79) all momenta $\mathbf{p}$ by $\mathbf{p}+\mathbf{K}/2$ and $-\mathbf{p}$ by $-\mathbf{p}+\mathbf{K}/2$ as well as $\tilde{\Omega}^m_{\mathbf{K}=0}$ by $\tilde{\Omega}^m_{\mathbf{K}}$.

In comparison with the usual m-wave equation (31), the derived Eq. (73) contains all polariton effects explicitly. This corresponds to the substitution of the x lines modified by the polariton effect instead of the unperturbed x lines (see Fig. 3). In the limit $p \gg \sqrt{\epsilon_0}\omega_t/c$, Eq. (73) reduces to Eq. (31). A natural correction of the absorption spectra of Eqs. (59) and (60) is thus the replace-

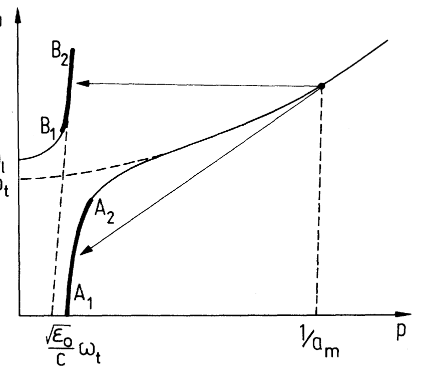

FIG. 5. Illustration of the optical decay of an m (see text).

ment of the unperturbed wave function $\Psi(\mathbf l)$ by $\tilde\Psi(\mathbf l)$, which includes polariton effects. For the 2γ m absorption such a correction cannot be treated by means of perturbation theory. Strong polariton modifications of the m-wave function $\Psi(\mathbf l)$ occur at small momenta $\hbar\mathbf l$ which belong to the optical range. In turn, for polaritons with momenta $\hbar\mathbf p$ and $\hbar\mathbf k$ of this range, an optical 2γ m formation takes place.

The analysis of Eq. (73) is a complex problem. At present we give only some comments. If only the lower polariton branch is taken into account, the initial Eq. (73) can be reduced to

$$(\omega_{\mathbf p}^- + \omega_{-\mathbf p}^- - 2\omega_t)\tilde\Psi(\mathbf p) + \phi^-(\mathbf p,\omega_{\mathbf p}^-)\phi^-(-\mathbf p,\omega_{-\mathbf p}^-) \times \sum_{\mathbf p'} W_{12}(\mathbf p-\mathbf p')\tilde\Psi(\mathbf p') = (\tilde\Omega^m_{\mathbf K=0} - 2\omega_t)\tilde\Psi(\mathbf p) . \quad (80)$$

The proper nonstationary solution $\tilde\Psi$ represents an outgoing spherical wave and corresponds to the radiative decay of an m state, i.e., $\tilde\Omega^m_{\mathbf K=0} = \Omega^m_{\mathbf K=0} - i\gamma^m_{\mathbf K=0}$. This value of the inverse lifetime of the m $\mathbf K=0$ has to be used in Eqs. (59) and (60) in order to get a self-consistent description of the absorption of two γ's with $\mathbf p$ and $\mathbf k = -\mathbf p$ with the initial Hamiltonian Eq. (29) or (38).

The radiative decay can be understood easily from Eq. (80) and is illustrated in Fig. 5. When the constituent x of the m acquires a small momentum within the optical range (the sectors A_1A_2 and B_1B_2 in Fig. 5), the corresponding m undergoes a radiative annihilation. In other words, the linear part of the lower polariton branch corresponds to the case without a bound m state. An m can exist as a well-defined excitation, if the optical range $p \simeq \omega_l\sqrt{\epsilon_0}/c$ is much smaller than the inverse m radius a_m^{-1}, i.e., if $\epsilon^m \gg \gamma^m_{\mathbf K=0}$. The radiative decay of an m is thus due to the polariton dispersion of its constituent x's. The decay is not a tunneling processes out of a metastable state.[35]

Both sets of Eqs. (33)–(35) and (40)–(42) formally give the same final equations for m luminescence, but give different results for m optical creation. We believe that this asymmetry is due to inconsistencies with respect to the internal x structure of the m in the phenomenological approach.

VI. CONCLUSIONS

In this work we have developed a self-consistent theory of the optical nonlinearities connected with the m state and compared it with the often-used phenomenological theory. The self-consistent approach is derived directly from the basic e-h-γ picture, taking as important ingredients the spin of the excitations as well as the proper quantum dependence of m on x explicitly into account. We have applied this concept to the description of the x-m optical Stark effect, to the 2γ m absorption, and to the m luminescence. For all these effects, the results of our self-consistent approach and the conventional one have been compared. The main conclusions are as follows.

(i) Instead of the phenomenological scenario of the 2γ creation of m ($\gamma+\gamma\rightarrow x+\gamma\rightarrow m$), the self-consistent description of the m absorption follows the scheme ($\gamma+\gamma\rightarrow x+x\rightarrow m$). In the phenomenological picture the m formation is described by the optical matrix element $M_1(\mathbf p,\mathbf k)$ related to the conversion of an x and a γ to an m. Here the two x's of the optically created m have the same momenta as the absorbed γ's. According to the Hopfield concept of polariton absorption, this does not describe true absorption. In the self-consistent approach the m is created by the Coulombic x-x scattering $M_2(\mathbf p,\mathbf k)$ of two x's with opposite internal spin structure. This process ensures a change of the initial γ momenta and breaks the quantum coherence between the γ's and the m. Even in the polariton representation these two approaches lead to the different results, both for the x-m optical Stark effect and for the 2γ m absorption.

(ii) The resonant 2γ m absorption coefficients $K_m^{(2)}(\omega,\omega_{\mathbf k})$ and $\tilde K_m^{(2)}(\omega,\omega_{\mathbf k})$ have been derived for the two approaches by taking into account the polariton character of the propagation of the two absorbed electromagnetic waves with frequencies $\omega_1=\omega$ and $\omega_2=\omega_{\mathbf k}$ and opposite circular polarizations. The absorption spectra of the self-consistent and the phenomenological theories coincide only if one of the frequencies ω_i is in exact resonance with the x line position ω_t, i.e., if $\omega_{1(2)}\rightarrow\omega_t$ and $\omega_{2(1)}\rightarrow\omega_t-\epsilon^m$. The differences between the self-consistent and the phenomenological approaches are large both around the degenerate 2γ absorption, where $\omega_1=\omega_2=\frac{1}{2}\Omega^m_{\mathbf K=0}$, and for strongly nondegenerate absorption, where $\omega_{1(2)}<\omega_t-\epsilon^m$ and $\omega_{2(1)}>\omega_t$. The self-consistent description of the m optical excitation in the direct-gap semiconductors can be tested experimentally.

(iii) A Schrödinger equation for the m-wave function $\tilde\Psi(\mathbf l,t)$ has been derived within a self-consistent approach which takes polariton effects into account explicitly. This equation allows us to describe the luminescence of an m. For a quantitative analysis of the 2γ absorption, the true m-wave function $\tilde\Psi(\mathbf l)$ which contains polariton effects has to be used.

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