Ultrafast Auger spectroscopy of quantum well excitons in a strong magnetic field

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We study theoretically the ultrafast nonlinear optical response of quantum well excitons in a perpendicular magnetic field. We address the role of many-body correlations originating from the electron scattering between Landau levels (LL). In the linear optical response, the processes involving inter-LL transitions are suppressed provided that the magnetic field is sufficiently strong. However, in the nonlinear response, the Auger processes involving inter-LL scattering of two photoexcited electrons remain unsuppressed. We show that Auger scattering plays the dominant role in the coherent exciton dynamics in strong magnetic field. We perform numerical calculations for the third-order four-wave-mixing (FWM) polarization which incorporate the Auger processes nonperturbatively. We find that inter-LL scattering leads to a strong enhancement and to oscillations of the FWM signal at negative time delays. These oscillations represent quantum beats between optically-inactive two-exciton states related to each other via Auger processes.

I. INTRODUCTION

It has been established that many-body processes play an important role in the transient optical response of semiconductors in the coherent regime[1, 2, 3, 4]. The Coulomb correlations between photoexcited carriers are especially strong in the presence of magnetic field. By suppressing kinetic energy of electrons and holes in two spatial dimensions (magnetic confinement), a high magnetic field enhances the relative strength of interactions between them[5, 6, 7, 8, 9, 10]. In bulk semiconductors, a dominant role of Coulomb correlations in magnetic field was demonstrated in four-wave mixing (FWM) spectroscopy experiments[12, 13, 14]. For example, a huge (several orders of magnitude) enhancement of the FWM signal was observed as the field exceeded certain characteristic value. A crossover to strongly-correlated regime occurs when the magnetic length, \( l \), becomes smaller than the excitonic Bohr radius, \( a_B \).

In quantum wells (QW) in perpendicular magnetic field, the energy spectrum is discrete so one would expect even stronger effect of interactions on the optical response. The linear absorption spectrum is dominated by a bound magnetoexciton (MX) state that incorporates electron and hole transitions between Landau levels (LL) in conduction and valence band, respectively. In strong field, such that the cyclotron energy, \( \hbar \omega_c \), is much larger than characteristic interaction energy, \( E_0 \sim e^2/\kappa l \) (here \( \kappa \) is the dielectric constant), the processes involving transitions between different LL’s are suppressed, and the lowest MX state is comprised of \( n = 0 \) LL electron-hole (e-h) pair with magnetic-field-dependent energy dispersion[15]. However, owing to the e-h symmetry for any given LL, such MX’s do not interact with each other due to a cancellation of Coulomb matrix elements between electrons and holes[16, 17]. For this reason, the nonlinear optical response of \( n = 0 \) MX’s is similar to that of noninteracting two-level systems[5, 6, 7] unless there is a sufficient e-h asymmetry because of, e. g., differing band offsets or disorder[18]. In the latter case, Coulomb correlations become important for both pump-probe[19] or FWM[21, 22] spectroscopy; however, in undoped QW’s, such an asymmetry is weak. In weaker magnetic field (\( \hbar \omega_c \leq E_0 \)), when LL mixing is strong, the coherent optical response in QW’s was studied in Hartree-Fock approximation (HA) within semiconductor Bloch equations technique[2, 3].

Here we study the role of many-body correlations in coherent optical spectroscopy of QW MX’s excited to upper \( (n > 0) \) LL’s. We focus on the case of a sufficiently strong magnetic field, \( \hbar \omega_c \gg E_0 \), so that individual optically-excited MX, with binding energy \( \sim E_0 \), is comprised of a single (e. g., \( n = 1 \)) LL e-h pair. For such fields, the processes involving inter-LL transitions do not contribute to linear response even if optical frequency is tuned to excite interband transitions at upper LL’s. However, as we demonstrate below, Coulomb correlations between e-h pairs excited to \( n \geq 1 \) LL’s are significant. Such correlations originate from Auger processes which involve inter-LL scattering of two photoexcited electrons. For example, two electrons on \( n = 1 \) LL can scatter to \( n = 0 \) and \( n = 2 \) LL’s, as illustrated in Fig. 1. Since this is a resonant process (LL’s are equidistant), it does not depend on the LL separation and, therefore, can take place even in a strong field, \( \hbar \omega_c/E_0 \gg 1 \). The inter-LL Auger processes has been previously observed in luminescence experiments[22].

We show that the Auger processes play the dominant role in the nonlinear spectroscopy of QW’s in strong magnetic field. Since they involve inter-LL scattering of charged carriers, the e-h symmetry no longer holds which gives rise to interactions between MX’s. It should be emphasized that since the LL’s are discrete, the amplitude of Auger process can be large and, in fact, is restricted mainly by the inhomogeneous[24, 25] or homogeneous (due to phonons) LL broadening. In fact, in strong field, the relevant energy scale, \( E_0 \sim e^2/\kappa l \), is set by interactions, so that an adequate
description of the nonlinear optical response should treat the Auger processes nonperturbatively. For example, the simple process described above (Fig. 1) is followed by Auger scattering of the electrons back to $n = 1$ LL — a process completely irrelevant in the luminescence. Such multiple Auger processes, by effectively causing interaction between MX's, give rise to coherent signature in the FWM spectroscopy. We perform numerical calculations of FWM polarization in strong field by including Auger-scattering exactly, in all orders of perturbation theory. We find a strong enhancement as well as oscillations of the FWM signal for negative time delays. These oscillations are identified as quantum beats originating from the interference between four-particle correlated states that are related to each other via Auger processes.

In Section II to we outline the formalism for evaluating the nonlinear polarization and compute the relevant matrix elements. In Section III we consider the contribution of the Auger processes into polarization. In Section IV we show the results of numerical calculations for the case of $n = 1$ LL. Section V concludes the paper.

II. EXCITON DYNAMICS ON ARBITRARY LANDAU LEVELS

A. General formalism

We consider a 2D system in strong perpendicular magnetic field with two-band Hamiltonian

$$H = H_0 + v(r),$$

where $H_0$ is free two-band Hamiltonian and $v(r)$ is the Coulomb potential. In the FWM spectroscopy, the sample is subjected to the probe and pump laser pulses, separated by time delay $\tau$, with electric field intensities $E_1(t)$ and $E_2(t)$ and wave vectors $k_1$ and $k_2$, respectively, and the signal along the direction $2k - k_1$ is measured. In order to obtain the third-order optical response, we adopt the formalism of Ref. [21] generalized to the case of arbitrary LL. The third-order FWM optical polarization has the form

$$P_{\text{FWM}}(t) = e^{i(2k_2 - k_1) \cdot \tau} P(t)$$

with

$$\hat{P}(t) = i\mu^2 e^{-i\omega_0 t} \int_{-\infty}^{t} dt' E_1^*(t') \times \left( \langle 0 | T e^{-iH(t-t')} T_{\text{FWM}}^\dagger(t')|0 \rangle - (t \leftrightarrow t') \right).$$

(1)

where $\mu$ is the the interband dipole matrix element, $\omega_0$ is the laser central frequency (we work in rotating frame), and $|0\rangle$ is the ground state of $H$. Here

$$T = \mu \sum_n U_n, \quad U_n = \sum_k b_{-kn} a_{kn},$$

(2)

is the interband transition operator ($a_{kn}$ and $b_{kn}$ are electron and hole annihilation operators, respectively), while the state $T_{\text{FWM}}^\dagger(t')|0\rangle$ stands for

$$T_{\text{FWM}}^\dagger|0\rangle = TW_0^\dagger|0\rangle - P_0^\dagger T P_0^\dagger|0\rangle,$$

(3)

where the single e-h pair state $P_0^\dagger(t)|0\rangle$ and the two e-h pair state $W_0^\dagger(t)|0\rangle$ satisfy the equations

$$i\partial_t P_0^\dagger(t)|0\rangle = H P_0^\dagger(t)|0\rangle + \mu \varepsilon_2(t) T^\dagger|0\rangle,$$

(4)
and
\[ i\partial_t W(t)|0\rangle = HW(t)|0\rangle + \mu \mathcal{E}_2(t)T^\dagger P(t)|0\rangle, \]
respectively; these equations describe the time-evolution (governed by the Hamiltonian \( H \)) of a single-exciton and two-exciton states [4].

\[\text{B. Basis}\]

\subsection{Single-exciton states}

The polarization is determined by the time-dependence of relevant one and two-pair states. In order to solve Eqs. (4,5), we use the standard basis for an electron in the Hilbert subspace with zero total momentum.

\[ \Psi_{\text{m}}(r_1, r_2) = N^{-1/2} \sum_k e^{-ikp_z t} \psi_{q/k, m}(r_1) \psi_{q/k, m}(r_2) = \frac{1}{L} e^{ip \cdot R - ixy/\ell^2} \varphi_{mn}(r + \ell^2 p \times z), \]

where \( p \) is the center-of-mass momentum of e-h pair, \( r = r_1 - r_2, R = (r_1 + r_2)/2 \) are the relative and the average coordinates, respectively, \( N = L^2/2\pi \ell^2 \) is the LL degeneracy, \( \ell \) is the unit vector in the direction of the magnetic field. Here, \( \psi_{kn}(r_1) \) and \( \psi_{-kn}(r_2) = \psi_{kn}^*(r_2) \) are the Landau wave-functions for electron and hole, and \( \varphi_{mn}(r) \) is given by

\[ \varphi_{mn}(z) = \sqrt{\frac{n!}{m!}} \left( \frac{iz}{\sqrt{2}} \right)^{m-n} L_n^{m-n} \left( \frac{|z|^2}{2\ell^2} \right) e^{-|z|^2/4\ell^2}, \quad m > n \]

and \( \varphi_{mn}(z) = \varphi_{nm}(z^*) \) for \( m < n \), where \( L_n^{a}(x) \) is the Laguerre polynomial and \( z = x + iy \) is the complex coordinate. Note also relations \( \varphi_{mn}^*(z) = \varphi_{nm}(-z) = (-1)^{m-n} \varphi_{nm}(z) = (-1)^{m-n} \varphi_{mn}(z^*) \).

In this basis, the single-pair amplitude,

\[ P_{mn}(q, t) = N^{-1/2} \langle q; mn | P^\dagger | 0 \rangle, \]

can be easily found from Eq. (4). The matrix elements of the Coulomb potential, \( v(r) \), have the form

\[ V_{mn, m'n'}(p) = \langle p; mn | v | p; m'n' \rangle = -i^2 \int \frac{dq}{2\pi} v_q e^{ip \cdot q} \varphi_{mn'}^*(q) \varphi_{mn'}(q), \]

where \( q = q_x + iq_y \) is the complex momentum and we used the identity

\[ \int dr \varphi_{mn}^*(r) e^{iq \cdot r} \varphi_{m'n'}(r) = 2\pi l^2 \varphi_{mn'}(q^*) \varphi_{mn'}(q). \]

From Eq. (4), we then obtain \( P_{mn}(q, t) = \delta_{qq'} \delta_{mn} P_{m}(t) \), where \( P_m(t) \) is the linear polarization, due to the pump, of the \( n \)th LL, satisfying

\[ (i\partial_t - \Omega_m) P_{m}(t) - \sum_n V_{mn, mn}(0) P_n(t) = \mu \mathcal{E}_2(t), \]

where \( \Omega_n = (n + 1/2) \omega_c + E_g - \omega_0 \) is the detuning (\( E_g \) is the bandgap).

\subsection{Two-exciton states}

Turning to the two-pair states, we introduce the amplitude

\[ W_{mn, m'n'}(p, t) = \langle p; mn, m'n' | W^\dagger | 0 \rangle, \]

where a complete orthogonal two-exciton basis in the Hilbert subspace with zero total momentum is constructed from symmetrized product of single-pair states:

\[ \Psi_{pnmn, m'n'}^{(2)}(r_1, r_2; r'_1, r'_2) = \frac{1}{2} \left[ \Psi_{pnmn}(r_1, r_2) \Psi_{-pnm'n'}(r'_1, r'_2) + \Psi_{pnmn}(r'_1, r'_2) \Psi_{-pnm'n'}(r_1, r_2) \right], \]
The corresponding matrix elements of Coulomb potential can be explicitly calculated as,

\[ \langle p; mn, m'n'|v| q; m_1n_1, m_1'n_1' \rangle = \delta_{pq} \left[ \delta_{mn_1} \delta_{nn_1} V_{m'n', m_1'n_1'}(p) + \delta_{m'n_1} \delta_{n'n_1} V_{mn, m_1n_1}(p) \right] + \frac{1}{L^2} V_{mn, m'n'; m_1n_1, m_1'n_1'}(p, q), \]  

(14)

where symmetrization with respect to \((mn) \leftrightarrow (m'n')\) is implicit hereafter. The last term describes MX-MX interaction accompanied by the momentum exchange.

\[ V_{mn, m'n'; m_1n_1, m_1'n_1'}(p, q) = 2\pi l^2 \int_{p-q} \left[ e^{-i\mathbf{q}\cdot \mathbf{r}/2} \varphi_{mn}(r - i\mathbf{q} \times \mathbf{z}/2) \varphi_{m'n'}(r + i\mathbf{q} \times \mathbf{z}/2) \right] = \delta_{mn'} \sqrt{2\pi l^2} \varphi_{mn'}(q), \]  

\[ \int d\mathbf{r} e^{i\mathbf{q}\cdot \mathbf{r}/2} \varphi_{mn}(r - i\mathbf{q} \times \mathbf{z}/2) \varphi_{m'n'}(r + i\mathbf{q} \times \mathbf{z}/2) = \delta_{nn'} \sqrt{2\pi l^2} \varphi_{mn'}(q'), \]  

(16)

Equation for two-MX amplitude \( W_{mn, m'n'; q, t} = \langle p; mn, m'n'|W^\dagger|0 \rangle \) is obtained by projecting Eq. (5) onto two-MX basis states,

\[ i\partial_t W_{mn, m'n'}(p, t) = \langle p; mn, m'n'|H|W^\dagger|0 \rangle + \mathcal{E}_2(t) \langle p; mn, m'n'|T^\dagger P^\dagger|0 \rangle, \]  

(17)

where the last (source) term can be found using Eq. (14),

\[ \langle p; mn, m'n'|T^\dagger P^\dagger|0 \rangle \]  

(18)

where we used the expansion

\[ U_m^\dagger U_n^\dagger|0 \rangle = N|0; mn, nn \rangle - \sum_p \langle p; mn, nn \rangle \]  

(19)

(the second term comes from the exchange).

3. FWM polarization

In the following we will be interested in the exciton-exciton interactions contribution into the FWM polarization. Correspondingly, we separate out the interactions-induced contribution by writing \( W^\dagger|0 \rangle = W_0^\dagger|0 \rangle + W_2^\dagger|0 \rangle \), where the first term, corresponding to non-interacting excitons, after being combined with the second term of (5), gives the Pauli blocking contribution of non-interacting excitons, and will be included in the numerical calculations below. The second term of the above decomposition gives the exciton-exciton interaction contribution to the polarization,

\[ \tilde{P}^{xx}(t) = i\mu^2 e^{-i\omega t} \int_{-\infty}^{\infty} dt' \mathcal{E}_1(t') \left[ \phi^{xx}(t, t') - \phi^{xx}(t', t) \right], \]  

(20)

with

\[ \phi^{xx}(t, t') = \langle 0| Te^{-iH(t-t')} TW^\dagger_{xx}|0 \rangle = \sum_{p, mn, m'n'} S_{mn, m'n'}(p, t-t') W_{mn, m'n'}(p, t'), \]  

(21)
where the function \( S_{mn,m'n'}(p,t) \equiv \langle 0|T e^{-i\hat{H}t}|p; mn, m'n' \rangle \) describes the propagation of an \( e-h \) pairs, created by pump and probe pulses, in the FWM direction, and can be expressed via the time-evolution operator \( K(t) \) as

\[
S_{mn,m'n'}(p,t) = \mu^2 \sum_{m_1} K_{m_1,m}(t) (N\delta_{p0}\delta_{mn}\delta_{m'n'} - \delta_{mn'}\delta_{m'n}),
\]

with the matrix elements \( K_{mn}(t) \) satisfying

\[
(i\partial_t - \Omega_m)K_{mn}(t) = \sum_{m_1} V_{mm_1,m_1n}(0)K_{m_1n}(t),
\]

and \( K_{mn}(0) = \delta_{mn} \). Putting all together, we obtain

\[
\phi^{xx}(t,t') = \mu^2 \sum_{m_1,mn} K_{m_1,m}(t-t')\chi_{mn}(t'),
\]

where

\[
\chi_{mn}(t) = W^{xx}_{mn,nn}(0,t) - \frac{1}{N} \sum_p W^{xx}_{mn,nn}(p,t).
\]

Substituting the above decomposition \( W_{mn,m'n'}(p,t) = W^0_{mn,m'n'}(p,t) + W^{xx}_{mn,m'n'}(p,t) \) into Eq. (17), where the non-interacting term has the form

\[
W^0_{mn,m'n'}(p,t) = \frac{1}{2} P_{mn}(t)P_{m'n'}(t)(N\delta_{p0}\delta_{mn}\delta_{m'n'} - \delta_{mn'}\delta_{m'n}),
\]

we obtain the following equation for \( W^{xx}_{mn,m'n'}(p,t) \):

\[
(i\partial_t - \Omega_m)W^{xx}_{mn,m'n'}(p,t) = \sum_{q,m_1,n_1'} \langle p; mn, m'n' | H | q; m_1n_1, m_1'n_1' \rangle W^{xx}_{m_1n_1,m_1'n_1'}(q,t) + J_{mn,m'n'}(p,t),
\]

where the matrix elements of the Hamiltonian were calculated above and the source term is given by

\[
J_{mn,m'n'}(p,t) = \sum_{q,m_1,n_1'} \langle p; mn, m'n' | H | q; m_1n_1, m_1'n_1' \rangle W^0_{m_1n_1,m_1'n_1'}(q,t)
- \frac{1}{2} \sum_{m_1} \left[ \Omega_{mm_1}P_{m_1}(t)P_{m'}(t) + \Omega_{m'm_1}P_{m_1}(t)P_{m'}(t) \right] 
\times (N\delta_{p0}\delta_{mn}\delta_{m'n'} - \delta_{mn'}\delta_{m'n}),
\]

with \( \Omega_{mn} = \Omega_m\delta_{mn} - V_{mm,nn} \). Substitution of Eqs. (11) and (26) into (28) yields a closed-form but rather complicated expression for \( J_{mn,m'n'}(p,t) \). In the following we will need to solve Eqs. (27) only for certain LL’s corresponding to the Auger processes.

### III. Auger Processes

We consider the case of strong magnetic field so that \( e^2/l \ll \omega_c \). In this case all single-pair processes involving inter-LL transitions are suppressed. For example, neglecting non-diagonal coupling, Eq. (11) simplifies to

\[
(i\partial_t - \Omega_n - V_{nn,nn}(0) + i\Gamma)P_n(t) = \mu\varepsilon_2(t),
\]

and similarly the single-pair evolution operator has only diagonal matrix elements, \( K_{mn}(t) = \delta_{mn}K_n(t) \) with

\[
K_n(t) = e^{-i(\Omega_n + V_{nn,nn}(0) - i\Gamma)t},
\]

where \( \Gamma \) is the MX homogeneous width.

In the case when two pairs are excited, inter-LL scattering becomes possible via Auger processes (see Fig. 1). Let the pump pulse, tuned to \( n \)th LL, excite two \( e-h \) pairs. Then the electrons can Auger-scatter with each other to \( n + \alpha \) and \( n - \alpha \) LL’s, and holes can Auger-scatter with each other to \( n + \beta \) and \( n - \beta \) LL’s. Since these are the only resonant
processes, all other inter-LL processes are suppressed. It is now convenient to introduce new notations which refer to the LL number to which the e-h pairs were initially excited,

\[ E^{\alpha\beta\gamma}_{\mathbf{n}\mathbf{p}} \equiv V_{n+n+\mathbf{n}\gamma\alpha\beta\gamma\alpha\beta}(\mathbf{p}) = -2\pi l^2 \int \frac{dq}{(2\pi)^2} e^{i\mathbf{p} \cdot \mathbf{q}} v_{\mathbf{q}} \varphi^{\alpha\gamma}_{\mathbf{n}}(\mathbf{q}) \varphi^{\beta\gamma}_{\mathbf{n}}(\mathbf{q}), \]  

(31)

and

\[ V^{\alpha,\beta,\alpha',\beta'}_{n}(\mathbf{p}, \mathbf{q}) = 2\pi l^2 v_{\mathbf{p} - \mathbf{q}} \left[ e^{-i\mathbf{p} \cdot \mathbf{q}^2} \varphi^{\alpha\gamma}_{\mathbf{n}}(\mathbf{p} - \mathbf{q}) \varphi^{\beta\gamma}_{\mathbf{n}}(\mathbf{p} - \mathbf{q}) (1 - \delta_{\alpha\alpha'}) \delta_{\beta\beta'} + e^{i\mathbf{p} \cdot \mathbf{q}^2} \varphi^{\beta\gamma}_{\mathbf{n}}(\mathbf{p} - \mathbf{q}) \varphi^{\alpha\gamma}_{\mathbf{n}}(\mathbf{p} - \mathbf{q}) (1 - \delta_{\beta\beta'}) \delta_{\alpha\alpha'} \right] \]

\[ -\delta_{\alpha\alpha'} \delta_{\beta\beta'} \left[ \varphi^{\alpha\gamma}_{\mathbf{n}}(\mathbf{p} - \mathbf{q}) \varphi^{\beta\gamma}_{\mathbf{n}}(\mathbf{p} - \mathbf{q}) + \varphi^{\alpha\gamma}_{\mathbf{n}}(\mathbf{p} - \mathbf{q}) \varphi^{\beta\gamma}_{\mathbf{n}}(\mathbf{p} - \mathbf{q}) \right], \]

(32)

where

\[ \varphi^{\alpha\beta}_{n}(p) \equiv \varphi^{\alpha\beta}_{n+n}(p) = \sqrt{\frac{(n + \beta)!}{(n + \alpha)!}} \left( \frac{ipl}{\sqrt{2}} \right)^{\alpha - \beta} L^{\alpha - \beta}_{n+\beta}(\frac{|p|^2 l^2 / 4}{\sqrt{2}\pi l^2}), \quad \alpha \geq \beta, \]

(33)

and \( \varphi^{\alpha\beta}_{n}(p) = \varphi^{\alpha\beta}_{n+n}(p) \) for \( \alpha < \beta \).

Since all non-Auger inter-LL transitions are suppressed, we restrict ourselves only with intermediate states related to each other via Auger processes. Then the relevant two-MX amplitudes \( w^{\alpha\beta}_{\mathbf{n}\mathbf{p}}(t) \equiv W^{\alpha\beta}_{n+n+n+n-n-n}(\mathbf{p}, \mathbf{t}) \) satisfy the following system

\[ [i\partial_t - 2\Omega_n E^{\alpha\beta}_{\mathbf{n}\mathbf{p}} - E^{-\alpha\beta}_{\mathbf{n}\mathbf{p}}] w^{\alpha\beta}_{\mathbf{n}\mathbf{p}}(t) = \sum_{\alpha',\beta'} \int \frac{dq}{(2\pi)^2} V^{\alpha,\beta,\alpha',\beta'}_{n}(\mathbf{p}, \mathbf{q}) w^{\alpha',\beta'}_{\mathbf{n}\mathbf{q}}(t) + m^{\alpha\beta}_{\mathbf{n}\mathbf{p}}(t). \]

(34)

Note that \( w^{\alpha\beta}_{\mathbf{n}\mathbf{p}}(t) = w^{-\alpha\beta}_{\mathbf{n}\mathbf{p}}(t) \). The source term \( m^{\alpha\beta}_{\mathbf{n}\mathbf{p}}(t) \) can be calculated from Eq. (28) as

\[ m^{\alpha\beta}_{\mathbf{n}\mathbf{p}}(t) = \left[ v_{\mathbf{p}} \varphi^{\alpha\beta}_{n+n}(p) \right] (1 - \delta_{\alpha\beta}) + E^{\alpha\beta}_{\mathbf{n}\mathbf{p}} - \epsilon^{\alpha\beta}_{\mathbf{n}\mathbf{p}} (1 - \delta_{\alpha\beta}) \]

\[ \times \left[ P_{n+n}(t) P_{n-n}(t) + P_{n+n}(t) P_{n-n}(t) \right], \]

(35)

where we used \( W^{\alpha\beta}_{n+n+n+n-n-n}(\mathbf{p}, \mathbf{t}) = \frac{1}{2} P_{n+n} P_{n-n} (N \delta_{\mathbf{p} \mathbf{p}} - \delta_{\mathbf{p} \mathbf{p}}) \). In the absence of Auger scattering \( \alpha = \beta = 0 \), we have \( w^{00}_{\mathbf{n}\mathbf{p}}(t) = 0 \), i.e. interactions do not contribute to the FWM signal (in the ideal 2D case).

Using that \( w^{\alpha\beta}_{\mathbf{n}\mathbf{p}}(t) \) is related to its Fourier transform as \( w^{\alpha\beta}_{\mathbf{n}\mathbf{p}}(t) = -N^{-1} \sum_{\mathbf{q}} e^{i\mathbf{q} \cdot \mathbf{p}^2} u^{\alpha\beta}_{\mathbf{n}\mathbf{q}}(t) \), we obtain from Eq. (28) that

\[ \chi^{\alpha\beta}_{n}(t) \equiv \chi^{n+n-n-n}(t) = 2w^{00}_{\mathbf{n}\mathbf{0}}(t). \]

Using Eqs. (30) and (30), the FWM polarization can be easily expressed in terms of amplitudes \( w^{\alpha\beta}_{\mathbf{n}\mathbf{0}}(t) \). In the following, we consider the case when the spectral width of the pulse is smaller than the LL separation. In this case, we have \( P_{n\pm\alpha} = \delta_{\alpha\alpha} P_{n}(t) \), and the FWM polarization takes the form (restoring Pauli blocking contribution)

\[ \tilde{P}(t) = \tilde{P}^{xx}(t) + \tilde{P}^{PB}(t), \]

(37)

with

\[ \tilde{P}^{xx}(t, \tau) = i\mu^2 \int_{-\infty}^{t} dt' e^{-i\Gamma(t'-t')-i\omega_{0}(t'+t')} \mathcal{E}_{1}(t') \left[ e^{-i(\Omega_{n+n} E^{n}_{\mathbf{n}\mathbf{0}})(t'-t')} \chi^{0}_{n}(t') - e^{i(\Omega_{n+n} E^{n}_{\mathbf{n}\mathbf{0}})(t'-t')} \chi^{0}_{n}(t') \right], \]

(38)

and

\[ \tilde{P}^{PB}(t, \tau) = i\mu^2 \int_{-\infty}^{t} dt' \mathcal{E}_{1}(t') e^{-i\omega_{0}(t'+t')} \left[ e^{-i\Gamma(t'-t')-i(\Omega_{n+n} E^{n}_{\mathbf{n}\mathbf{0}})(t'-t')} P_{n}^{2}(t') - P_{n}(t) P_{n}(t') \right] \]

\[ + \mu^3 P_{n}(t) \int_{-\infty}^{t} dt'' \mathcal{E}_{2}(t) \int_{-\infty}^{t} dt' e^{i\omega_{0}(t''+t')-i(\Omega_{n+n} E^{n}_{\mathbf{n}\mathbf{0}})(t'-t')-i\Gamma(t'-t')}, \]

(39)

where \( E^{n}_{\mathbf{n}\mathbf{0}} \equiv V_{n+n,n+n}(0) \) and

\[ P_{n}(t) = -i\mu \int_{-\infty}^{t} dt' \mathcal{E}_{2}(t') e^{-i(\Omega_{n+n} E_{n})(t'-t')-i\Gamma(t'-t')} \]

(40)

is the pump-induced polarization on nth LL.
IV. DISCUSSION AND NUMERICAL RESULTS

We consider the case when the central laser frequency is tuned to interband transition between \( n = 1 \) LL's. In this case, two exciton excited by the pump to \( n = 1 \) LL, can Auger-scatter only to \( n = 0 \) and \( n = 2 \) LL's, i.e., \( \alpha, \beta = 0, \pm 1 \) (see Fig. 1). System (31) then takes the form:

\[
(i\partial_t - 2\Omega_1 - 2E_{1p}^{(0)}u_{1p}^{(0)}(t) - \int \frac{dq}{(2\pi)^2} V_{11}^{(0)}(p, q)w_{1q}^{(0)}(t) - 2\int \frac{dq}{(2\pi)^2} \tilde{V}_{11}^{(0)}(p, q)w_{1q}^{(0)}(t) = 0,
\]

\[
(i\partial_t - 2\Omega_1 - E_{1p}^{(1)} - E_{1p}^{(1-1)} - \int \frac{dq}{(2\pi)^2} V_{11}^{(1)}(p, q)w_{1q}^{(1)}(t) - \int \frac{dq}{(2\pi)^2} \tilde{V}_{11}^{(1)}(p, q)w_{1q}^{(1)}(t) = 0,
\]

\[
(i\partial_t - 2\Omega_1 - 2E_{1p}^{(1)} - E_{1p}^{(1-1)} - \int \frac{dq}{(2\pi)^2} V_{11}^{(1-1)}(p, q)w_{1q}^{(1-1)}(t) - \int \frac{dq}{(2\pi)^2} \tilde{V}_{11}^{(1-1)}(p, q)w_{1q}^{(1-1)}(t) = 0,
\]

\[
(i\partial_t - 2\Omega_1 - E_{1p}^{(1+1)} - E_{1p}^{(1+1-1)} - \int \frac{dq}{(2\pi)^2} V_{11}^{(1+1)}(p, q)w_{1q}^{(1+1)}(t) - \int \frac{dq}{(2\pi)^2} \tilde{V}_{11}^{(1+1)}(p, q)w_{1q}^{(1+1)}(t) + w_{11}^{(1)}(t) + w_{11}^{(1-1)}(t) = m_{1p}(t),
\]

where the source term and inter-pair Coulomb matrix elements are given by

\[
m_{1p}^{(1)}(t) = \left[ \frac{v_p}{2\pi l^2} e^{-p^2t^2/2} \left( \frac{p}{2} \right)^2 L_1(p^2t^2/2) - E_{1p}^{(1)} \right] \frac{P_2^2(t)}{2},
\]

\[
V_{11}^{(0)}(p, q) = -4\sin^2(p \times q) e^{-p^2q^2/2} \left( L_1(|p - q|^2t^2/2) \right)^2,
\]

\[
V_{11}^{(1)}(p, q) = -4\sin^2(p \times q) e^{-p^2q^2/2} \left( L_2(|p - q|^2t^2/2) \right),
\]

\[
V_{11}^{(1-1)}(p, q) = -4\sin^2(p \times q) e^{-p^2q^2/2} \left( L_1(|p - q|^2t^2/2) - L_2(|p - q|^2t^2/2) \right),
\]

\[
V_{11}^{(1+1)}(p, q) = 2\cos(p \times q) e^{-p^2q^2/2} \left( \frac{|p - q|^2t^2}{4} L_1(|p - q|^2t^2/2) \right),
\]

with \( v_q = 2\pi e^2/q \), while the single-exciton energies \( E_{1p}^{\alpha\beta} = E_{1p}^{\alpha\beta\alpha\beta} \) have the following form

\[
E_{1p}^{(0)} = -\sqrt{\frac{\pi}{2}} e^2 l \left[ \Phi \left( \frac{1}{2}, 1, -\frac{p^2t^2}{2} \right) - \Phi \left( \frac{3}{2}, 1, -\frac{p^2t^2}{2} \right) + \frac{3}{4} \Phi \left( \frac{5}{2}, 1, -\frac{p^2t^2}{2} \right) \right],
\]

\[
E_{1p}^{(1-1)} = -\sqrt{\frac{\pi}{2}} e^2 l \Phi \left( \frac{1}{2}, 1, -\frac{p^2t^2}{2} \right),
\]

\[
E_{1p}^{(1+1)} = -\sqrt{\frac{\pi}{2}} e^2 l \left[ \Phi \left( \frac{1}{2}, 1, -\frac{p^2t^2}{2} \right) - 2\Phi \left( \frac{3}{2}, 1, -\frac{p^2t^2}{2} \right) + \frac{15}{4} \Phi \left( \frac{5}{2}, 1, -\frac{p^2t^2}{2} \right) \right]
\]

\[
E_{1p}^{(1)} = -\sqrt{\pi} e^2 l \left[ \Phi \left( \frac{1}{2}, 1, -\frac{p^2t^2}{2} \right) - \frac{3}{2} \Phi \left( \frac{3}{2}, 1, -\frac{p^2t^2}{2} \right) + \frac{15}{8} \Phi \left( \frac{5}{2}, 1, -\frac{p^2t^2}{2} \right) \right]
\]

\[
E_{1p}^{(0-1)} = -\sqrt{\frac{\pi}{2}} e^2 l \left[ \Phi \left( \frac{1}{2}, 1, -\frac{p^2t^2}{2} \right) - \frac{1}{2} \Phi \left( \frac{3}{2}, 1, -\frac{p^2t^2}{2} \right) \right],
\]
\[
E^{1-1}_{1p} = -\sqrt{\frac{\pi e^2}{2 \ell}} \left[ \Phi \left( \frac{1}{2}, 1, \frac{-p^2 \ell^2}{2} \right) - \Phi \left( \frac{3}{2}, 1, \frac{-p^2 \ell^2}{2} \right) + \frac{3}{8} \Phi \left( \frac{5}{2}, 1, \frac{-p^2 \ell^2}{2} \right) \right],
\]

\[
\hat{E}^{01}_{1p} = -\sqrt{\frac{\pi e^2}{2 \ell}} \left[ 2\Phi \left( \frac{3}{2}, 1, \frac{-p^2 \ell^2}{2} \right) - \frac{3}{2} \Phi \left( \frac{5}{2}, 1, \frac{-p^2 \ell^2}{2} \right) \right],
\]

\(\Phi(a, b, z)\) being the confluent hypergeometric function, and the pump-induced polarization is given by

\[
P_1(t) = -i\mu \int_{-\infty}^{t} dt' e^{-i(O_1 + E^{00}_{10} - i\Gamma)(t-t')} E_2(t').
\]  \(43\)

The first equation in systems \(41\) describes the time-evolution of two \(e-h\) pairs, excited to \(n = 1 LL\) by the pump pulse [Fig. 1(a)], with and without inter-LL scattering (second and third terms, respectively). The third and second equations in \(41\) describe the similar time evolution of the state (c) in Fig. 1 and of the state obtained from (c) by exchanging the holes in the valence band. The last terms of the first three equations in \(41\) describe the Auger scattering that relates the corresponding states to the state (b) in Fig. 1 described by the fourth equation. Note that only state (b) lacks the \(e-h\) symmetry, i.e., does not transform into itself under replacement of electrons by holes and vice-versa in Fig. 1 as indicated by the source term in the rhs of the forth equation in \(41\). Its amplitude, \(w_{1q}^0(t)\), plays the role of the source for the rest of the system \(41\) (last terms) and thus represents the sole source for the interaction-induced polarization.

In the numerical calculations below, we consider resonant excitation, i.e., central frequency tuned at the transitions between ground state and \(n = 1 MX\) with binding energy

\[
E^{00}_{10} = -\frac{3}{4}E_0, \quad E_0 = \frac{\sqrt{\pi e^2}}{2 \ell},
\]  \(44\)

where \(E_0\) is the binding energy of the \(n = 0 MX\).

The above equations for \(w\) have been solved numerically using fourth-order Runge-Kutte routine. FWM polarization was then calculated from Eqs. 37-39 with \(n = 1\), for Gaussian pump and probe pulses with the duration \(t_p\) separated by time delay \(\tau\). Dephasing due to electron-phonon interactions have been accounted for by MX width \(\Gamma\) and by the width \(\gamma\) characterizing the dephasing of two-pair system. We emphasize that in the case of interacting excitons, \(\gamma\) does not necessarily equal \(2\Gamma\), as has been pointed out in experiment \(13\).

In Fig. 2 we show calculated time-integrated FWM signal,

\[
TI - FWM = \int dt|\langle P(t)\rangle|^2,
\]  \(45\)

for parameter values \(t_p = \hbar/E_0, \Gamma = 0.1E_0, \gamma = 0.05E_0\) versus time delay (in units of \(E_0\)). The dot-dashed curve corresponds to Pauli blocking contribution that comes from non-interacting excitons. It should be emphasized that, in strong magnetic field, the \(only\) interaction-induced contribution comes from Auger scattering, so the Hartree-Fock contribution is suppressed by a small parameter \(E_0/\hbar\omega_c < 1\). It can be seen that Auger scattering of magnetoexcitons strongly enhances the amplitude of FWM signal. For negative time delays, TI-FWM signal develops an exponential (with decay-time \(\hbar/\gamma\)) tail that is characteristic for interacting excitons. Furthermore, TI-FWM signal exhibits two sets of oscillations superimposed on each other. These oscillations represent, in fact, quantum beats between four-particle configurations contributing to FWM polarization. We identify more the pronounced oscillations with the interference between states excited state (a) and state (b) in Fig. 1 related to (a) by electron Auger-scattering in conduction band. These states are characterized by Coulomb energies [see Eqs. 41 and 43] \(E_a = 2E^{00}_{10} = -\frac{3}{4}E_0\) and \(E_b = E^{00}_{10} + E^{0-1}_{10} = -\frac{5}{4}E_0\), so the oscillations period corresponds to the energy difference \(E_b - E_a = \frac{1}{4}E_0\). The weaker oscillations originate from the interference between states (a) and (c) in Fig. 1 where the state (c) is characterized by energy \(E_c = 2E^{1-1}_{10} = -\frac{3}{4}E_0\), so the period is determined by \(E_c - E_a = \frac{1}{2}E_0\). Note that the state (d), obtained from (c) by hole exchange in the valence band, is characterized by the energy \(E_d = E^{11}_{10} + E^{-1-1}_{10} = -\frac{105}{64}E_0\) and the corresponding period, determined by \(E_d - E_a = \frac{9}{16}E_0\), is too long to be noticeable in Fig. 2. It should be emphasized, however, that the above estimates involve zero-momentum energies of each of the pairs that constitute four-particle correlated state. In fact, Auger processes are accompanied by a momentum exchange between MX’s, so only the total momentum of a four particle correlation is zero. This momentum exchange leads to the damping of oscillations. Since (a) and (c) are related via two Auger processes (in conduction and in valence band) the additional momentum exchange during the hole Auger-scattering in valence band leads to a much more stronger damping of (a)-(c) oscillations as compared to (a)-(b) oscillations.
FIG. 2: Calculated TI-FWM signal versus time delay (in units of magnetoeexciton energy) shows oscillations corresponding to the interference between states (a) and (b), and between states (a) and (c) in Fig. 1.

V. CONCLUSIONS

We investigated the role of inter-LL transitions in the coherent dynamics of quantum well excitons in strong magnetic field. While on the lowest LL, the suppression of inter-LL transitions results in the absence of exciton-exciton interactions, on higher LL levels the interactions become dominant due to resonant Auger processes. The coherent signature of exciton Auger-scattering can be traced in the FWM polarization as quantum beats corresponding to the interference between optically-inactive four-particle correlated states.

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