Collective oscillations driven by correlation in the nonlinear optical regime

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We present an analytical and numerical study of the coherent exciton polarization including exciton-exciton correlation. The time evolution after excitation with ultrashort optical pulses can be divided into a slowly varying polarization component and novel ultrafast collective modes. The frequency and damping of the collective modes are determined by the high-frequency properties of the retarded two-exciton correlation function, which includes Coulomb effects beyond the mean-field approximation. The overall time evolution depends on the low-frequency spectral behavior. The collective mode, well separated from the slower coherent density evolution, manifests itself in the coherent emission of a resonantly excited excitonic system, as demonstrated numerically.

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Observations of coherent optical phenomena require a sufficiently long dephasing time. Thus, coherent processes such as the Rabi oscillations and form-invariant pulse propagation (also known as self-induced transparency) are well studied both theoretically and experimentally in atomic vapors. In solid-state physics, the nonlinear optical properties of semiconductors are expected to exhibit analogous behavior, especially under resonant excitation of excitons, where the exciton transition approximates an ideal two-level system. However, there is a distinct difference in semiconductors. At moderate excitation densities, the mobile, large-radius Wannier excitons suffer strong Coulomb interaction on moderate excitation densities, the mobile, large-radius Wannier excitons suffer strong Coulomb interaction on moderate excitation densities, the mobile, large-radius Wannier excitons suffer strong Coulomb interaction on moderate excitation densities.

A mean-field theory of the exciton-exciton interaction leads to a purely coherent repulsive interaction, which does not yield a mechanism for dephasing due to the fluctuating effective local field. This level of approximation allows for nonlinear electric field-driven density oscillations, which are termed as Rabi oscillations. A further low-density approximate relation for the density and the polarization leads to a Gross-Pitaevskii equation for the exciton dynamics. This yields form-invariant soliton solutions for spatial variation. It is then exact to the third order in the exciting field. The whole equation is thus described by a nonlinear equation of motion for the polarization density $P$ of the form:

$$i\dot{P} = \left(\delta + \frac{\beta}{2}|P|^2\right)P - \frac{1}{2}\left(1 - \frac{|P|^2}{n_c}\right)\Omega + \frac{iP^*}{2}\int_{-\infty}^{t} F(t - t')P(t')^2dt'.$$

The equation is easily generalized to include the spin, higher exciton levels, and the continuum and to account for spatial variation. It is then exact to the third order in the exciting field. The whole equation is thus valid for the low-density regime. Our purpose here is to explore the temporal variation of the exciton dynamics at low excitation density and, therefore, we retain only the resonantly excited exciton transition and set $P$ to be the usual polarization density divided by $\mu|\phi_{1s}(0)|^2$, $\mu$ being the dipole moment and $\phi_{1s}(r)$ the relative motion wave function. For the terms in order on the right side of the equation, $\delta$ denotes the detuning of the central laser frequency from the resonance, $\beta > 0$ is a measure of the mean-field exciton-exciton interaction, and $n_c$ the maximum density divided by $|\phi_{1s}(0)|^2$ for the validity limit in the Pauli blocking term. The Rabi frequency $\Omega$ is given by the dipole energy of the laser field amplitude. Exciton-exciton correlation is contained in the retarded memory function $F(\tau)$. Similar effective equa-
tions without the correlation term \( F \) were already used in the literature. A simple way to account for the correlation effect by connecting the polarization equation to an antiparallel-spin biexciton density is equivalent to setting \( F \) to be the single-frequency biexciton propagator.

The retarded memory function is given by

\[
F(\tau) = V|\phi_{1s}(0)|^2\langle 0|D e^{-iH\tau}D|0\rangle \Theta(\tau),
\]

in terms of the Hamiltonian of the system \( H \), volume \( V \), and the interaction operator between two excitons,

\[
D = \int dr_1 \int dr_2 \int dr'_1 \int dr'_2 B(r_1, r_2) [v(r_1 - r'_1)] + v(r_2 - r'_2) - v(r_1 - r'_2) - v(r_2 - r'_1)] B(r'_1, r'_2),
\]

with the Coulomb interaction \( v(r) \) and the exciton annihilator

\[
B(r_1, r_2) = \phi_{1s}(r_1 - r_2) \psi_1^\dagger(r_1) \psi_2(r_2),
\]

where \( \psi_1^\dagger, \psi_2 \) are, respectively, the electron creator in the valence band and the annihilator in the conduction band. The formulas in configuration space show clearly the inclusion of the electron-hole attraction in each exciton, leaving only the interaction between the constituents of the two excitons in the interaction operator \( D \). The correlation effects contained in the memory function are in second or higher order of the interaction between two excitons.

Our numerical simulation below shows that intrinsic dephasing times due to exciton-exciton scatterings are of the order of a few picoseconds in a GaAs quantum well, of the same order as that due to electron-phonon. Thus, the conclusions drawn from the exclusion of the extrinsic dephasing would not be qualitatively changed by its inclusion. The equation for the polarization density resembles the Gross-Pitaevskii equation for a single component Bose-Einstein condensate. However, there are two essential differences, (1) the exciton coherence is driven externally, and (2) the effective interaction between polarization includes the correlation term beyond the mean-field theory, which may well be applied to the trapped atoms as their density increases.

After excitation with a short laser pulse at time \( t = 0 \), the coherent polarization evolves without the driving field \( \Omega \). We present first an analytic solution of Eq. (1) under the assumption of a sufficient slowly varying polarization \( P_0(t) \), which is then used to understand the behavior of the numerical solutions without this approximation. Under the slowly varying assumption, the rapidly decaying memory function can be decoupled from the polarization

\[
\int_{-\infty}^t F(t - t') P_0^2(t') dt' \approx P_0^2(t) \int_{0}^t F(t - \tau) d\tau.
\]

This is similar to a Markov approximation for the polarization but the scattering rate is still a time-dependent memory function, which we group with the mean-field term from Eq. (1) in a new function:

\[
g(t) = \int_{0}^t F(\tau) dt + i\beta.
\]

After the exciting pulse dies down, the detuning \( \delta \) term can be included in \( P_0 \) and removed from the equation of motion. For the resultant equation, the ansatz \( P_0 = n^{1/2} e^{i\phi} \) solves the slow time dynamics:

\[
n(t) = n_0 \left( 1 + n_0 \int_{0}^t g_R(t') dt' \right)^{-1},
\]

\[
\phi(t) = \phi_0 - \frac{1}{2} \int_{0}^t g_I(t') n(t') dt',
\]

for the initial conditions \( n(0) = n_0, \phi(0) = \phi_0 \) set by the exciting pulse. The real part of \( g(t) \), \( g_R(t) \), determines the dephasing of the coherent polarization and, thus, the exciton population. The imaginary part, \( g_I(t) \) determines the phase of the polarization. The mean-field effects are included in the total memory function \( g(t) \).

The normalized spectral density of the \( D-D \) correlation function, denoted by \( \rho(\omega) \), is \( 1/\pi \) times the real part of the Fourier transform of \( F(\tau)/F(0) \). The sum rule,

\[
F(0) \int_{0}^{\infty} \omega^{-1} \rho(\omega) d\omega = \beta
\]

shows important cancellation in \( g(t) \) between the mean-field exchange and the correlation effects. The low-frequency behavior of the spectral density is governed by a power law, \( \rho(\omega) \sim \omega^\alpha \), discussed below. It determines the asymptotic long-time dynamics via:

\[
g(t) \sim i \int_{0}^{\infty} \omega^{\alpha-1} e^{-i\omega t} d\omega = i^{\alpha+1} \Gamma(\alpha)t^{-\alpha},
\]

FIG. 1. Equal-spin correlation function (solid-line) for a one-dimensional model system. The inset shows the exact spectral density \( \rho(\omega) /\omega \) for small \( \omega \), indicating a linear dependence on \( \omega \).
where $\Gamma$ is the usual Gamma function, leading to a crossover from a decaying coherence for $\alpha \leq 1$ to a constant density $n(\infty) \leq n_0$ for $\alpha > 1$. In Figure 1, the spectral density of the equal-spin correlation function is plotted for a one-dimensional system defined in [13] with the exciton binding energy $\omega_x = 6.7$ meV, the one-electron bandwidth of 50 meV and equal electron and hole mass. An artificial broadening of 0.4 meV is used to smooth the energy splittings due to the finite simulation size of $N = 320$ sites. The inset shows the exact behavior of the spectral density for small $\omega$, indicating a linear behavior, i.e., $\alpha = 1$.

![Figure 1](image1.png)

**Figure 1.** The spectral density of the equal-spin correlation function is plotted for a one-dimensional system defined in [13] with the exciton binding energy $\omega_x = 6.7$ meV, the one-electron bandwidth of 50 meV and equal electron and hole mass. An artificial broadening of 0.4 meV is used to smooth the energy splittings due to the finite simulation size of $N = 320$ sites. The inset shows the exact behavior of the spectral density for small $\omega$, indicating a linear behavior, i.e., $\alpha = 1$.

$\rho(\omega) = \frac{\omega^\alpha \exp(-\omega/\omega_F)}{\Gamma(\alpha + 1)\omega_F^{\alpha+1}},$ (10)

for positive frequencies with a single parameter $\alpha(>0)$. The values for $\beta = 52\omega_x/3$, $1/n_c = 7$, and $F(0) \approx 14\omega_x^2$ are computed from the two-band model. The frequency scale $\omega_F = F(0)/\beta\alpha$ is fixed by the sum-rule [5].

![Figure 2](image2.png)

**Figure 2.** Coherent exciton density for the one-dimensional model of Fig. 1 for a pulse intensity FWHM of 200 fs and increasing pulse area $\Theta = \int \Omega(\tau)d\tau$. The time unit $T_x = 2\pi/\omega_x$.

**Figure 3.** Coherent polarization for the three-dimensional semiconductor model with $\omega_x = 4.2$ meV, pulse width 200 fs and pulse area $4\pi$ for various $\alpha$ of the model correlation function. The other parameters are given in the text. The dashed-dotted line is the excitation pulse.

Figure 3 shows the result of the numerical solution of Eq. (1) for the model correlation with various values for the parameter $\alpha$ for an excitation with a 200 fs laser pulse and area $4\pi$. For $\alpha \geq 2$, the asymptotic behavior of the exciton density changes to constant from decaying. For $\alpha \geq 1$, small oscillations are superimposed on the slow density evolution. The period of oscillations is about 200 fs for $\alpha = 4$. These oscillations do not correspond to the frequency peak $\omega_c = F(0)/\beta$ of the spectral density, which does not vary with increasing $\alpha$ as the peak narrows. They are collective modes in density fluctuation excited by the strong $4\pi$-pulse. They do not occur in the slowly-varying approximation [13]. It is, however, possible to treat the modes as a small, fast-varying perturbation $P_1$ to the slow component $P_0$. Inserting the combination $P = P_0 + P_1$ in Eq. (1) and keeping $P_1$ only to first order, we find

$$P_1 = -i\beta P_1 \left[ i\beta P_1 + \int_0^t F(t - t')P_1(t')dt' \right],$$

$$-\frac{P_0^2}{2} g(t)P_0^*(t).$$

The last contribution of Eq. (11), being $P_0P_1^*/P_0^*$ is negligible compared with $P_1$. The resulting linear equation can be solved by Fourier transform of $P_1(t)$ for a slowly-varying density to yield the collective-mode frequencies as zeros of the equation.
\[
\omega = n \Sigma(\omega),
\]
\[
\Sigma(\omega) = \beta - F(0) \int_0^\infty \frac{\rho(\omega') d\omega'}{\omega' - \omega - i0^+}.
\]  

Figure 3 shows a graphical solution of the collective mode equation from the crossings between the real part of \( \Sigma \) and the straight line \( \omega/n \). The \( \omega = 0 \) solution corresponds to the Goldstone mode \([14]\). Models for a range of \( \alpha \) parameters are shown. Above a critical density, e.g., \( n \approx 0.1 \) (in the density units of Fig. 3) for \( \alpha = 2 \), additional solutions with two high frequencies exist, but only the higher frequency solution with the weaker damping corresponds to the oscillating mode in Fig. 3.

The imaginary part of \( \Sigma \) represents the damping of the collective mode by the two-exciton continuum. It peaks around \( \omega = 2\omega_x \) where the real part rises steeply and, thus, damps out the lower frequency solution. We have not included the damping by two excitons when one (or both) of them is dissociated into an electron-hole pair in our computation but this would not qualitatively change the damping continuum. In the three-dimensional model where \( T_x \approx 1 \) ps, these oscillations have periods between 50–200 fs. The curvature in the \( \alpha = 0.5 \) case is exceptional, resulting in only one high-frequency solution whose damping goes down with increasing excitation density.

In conclusion, we extended the Gross-Pitaevskii equation for the exciton polarization at low densities to include correlation, which is an important counterpart to the mean-field exchange term and which provides an intrinsic dephasing mechanism. A consequence is the existence of collective oscillations riding on a slowly varying density. The Goldstone mode, though undetectable, indicates a Bose-Einstein condensate behavior, induced by optical excitation but sustained by exciton interaction.

Finding by nonlinear optical measurement of exciton polarization of the high-frequency mode, predicted by the same dispersion relation as the Goldstone mode, could then be argued to present evidence of condensation.

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