Evidence of Nonperturbative Continuum Correlations in Two-Dimensional Exciton Systems in Semiconductor Microcavities

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In nonlinear semiconductor optics, two-particle scattering is often being modeled successfully within the second Born approximation (2nd BA) of the Coulomb interaction. It is shown in this paper that, at low energies, such a perturbative treatment of Coulomb correlations applied to exciton-exciton scattering in *two*-dimensional systems fails even *qualitatively* (unless phenomenological or self-consistent dephasing processes are included in the theory). We show that the failure of the 2nd BA in two dimensions can be inferred from a comparison of our theoretical results with reported experiments of four-wave mixing signals from semiconductor microcavities.

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Linear excitonic absorption effects (see, e.g., [1]) and optical nonlinearities arising from exciton-exciton interactions in quasi-one-, two-, and three-dimensional semiconductors are well established (see, e.g., [2–4]). While the dimensionality dependence of *linear* excitonic effects is well understood (the simplest example being the increase of exciton binding energy with decreasing dimensionality), that of *nonlinear* excitonic effects is in general not clearly established. In this Letter, we discuss one aspect of this issue which concerns exciton-exciton scattering at low kinetic energies. Such scattering processes are commonly treated within the second Born approximation (2nd BA), because this approximation can already account, at least qualitatively, for all observable signatures of exciton-exciton scattering in nonlinear optical experiments. Moreover, numerous experiment-theory comparisons (including cases involving exciton-free-carrier scatterings) seem to validate the 2nd BA. Examples include the line shape of optical gain spectra in semiconductor lasers [5], photoluminescence from semiconductor quantum wells [6], and optical Stark shifts in semiconductor quantum wells [7].

While we do not dispute the validity of the model assumptions underlying those experiment-theory comparisons, we will show in the following that the 2nd BA in *two* dimensions should be used with caution. In fact, if the theoretical results are not smoothened by phenomenological or self-consistently calculated [6] dephasing rates, the 2nd BA T matrix (or scattering amplitude) governing low-energy exciton-exciton scattering would *diverge* in the limit of zero exciton center-of-mass-motion energies, while the exact scattering amplitude is known to *vanish* as $1/ln(energy)$ in the same limit [8]. These behaviors result from the dimensionality of the system and hold for any generic short-ranged potential independent of strength and other details. In analyzing semiconductor experiments, including the ones referenced above, dephasing introduced into the theory regularizes these nonanalytic behaviors, but their presence is still indicated by the large deviation of the 2nd BA T matrix from the exact one even for only moderately strong potentials (below, we will show this in more detail, cf. Fig. 2). Admittedly, since this deviation is quantitative instead of qualitative, its effects may not be ascertainable in many cases where other theoretical and/or experimental uncertainties are present. The 2nd BA theory would then be deemed satisfactory. Given this situation, one might ask if this theoretical breakdown of the 2nd BA is relevant at all to the interpretation of nonlinear optics experiments on two-dimensional semiconductor structures. It is the purpose of this paper (i) to show that third-order nonlinear experiments are direct probes of the exciton-exciton zero-momentum *off*-energy-shell T matrix and relate this special case to the general results of two-dimensional scattering theory, and (ii) to show that there indeed exist experimental data that yield signatures of the breakdown of the 2nd BA in two-dimensional exciton-exciton scatterings.

There have been other theoretical assessments of the limitations of the 2nd BA for carrier-carrier and carrier-phonon scatterings in semiconductors [9–11]. In particular, Ref. [11] also pointed out the relation between the breakdown of the 2nd BA for carrier-carrier scattering and the system's dimensionality. To our knowledge, these findings have not been corroborated by experiments.

In principle, the 2nd BA for exciton-exciton scattering fails in any $\chi^{(3)}$ measurement configuration on thin quantum wells. We choose here to demonstrate this failure within a microscopic theory of frequency-resolved degenerate four-wave mixing (FWM) because, when the quantum well is embedded in a microcavity, the FWM signals are particularly sensitive to higher-order exciton continuum correlations. We show below that the microcavity FWM data reported in Ref. [12] are indeed sufficiently sensitive to yield experimental indications of the failure of the 2nd BA in two dimensions. Although the two-dimensional 2nd BA, in principle, also fails in other physical systems, scattering experiments in two dimensions are generally more difficult to set up than in the exciton system in a quantum well microcavity.

In the following, we review and discuss our theoretical approach to four-wave mixing in semiconductor quantum wells in the 3rd-order nonlinear optical regime and its relationship to the quantum mechanics of two-particle scattering. Our theoretical approach is based on the dynamics-controlled truncation (DCT) formalism [13] and includes a quantitative evaluation for optical excitation frequencies close to the heavy-hole exciton resonance in thin quantum wells.

We start with the $\chi^{(3)}$-DCT equations [13] for the interband polarization and the coherent two-electron-two-hole (biexcitonic) correlation function (see, e.g., [14,15]). Following Ref. [16], we expand the equations in the exciton eigenfunction basis, taking into account the antisymmetry of the biexcitonic correlation function in the two-electron and the two-hole coordinates, and truncate to the $1s$ subspace. We evaluate the theory in the cw (continuous wave) configuration, with spectrally degenerate pump $[E_p(\omega)]$ and probe $[E_t(\omega)]$ light fields. In this case, the 3rd-order polarization can be written as

$$P_{\pm}^{(3)}(\omega) = [\chi^{++}(\omega)E_{p\pm}(\omega)E_{t\pm}^{*}(\omega) + \chi^{+-}(\omega)E_{p\mp}(\omega)E_{t\mp}^{*}(\omega)]E_{p\pm}(\omega), \quad (1)$$

where $+, -$ denote the circular polarization states, and the 3rd-order susceptibilities are given by

$$\left\{ \begin{matrix} \chi^{++}(\omega) \\ \chi^{+-}(\omega) \end{matrix} \right\} = -\frac{C}{[\hbar\omega - \varepsilon(\vec{0}) + i\gamma_2]^2[(\hbar\omega - \varepsilon(\vec{0}))^2 + \gamma_2^2]} \left\{ \begin{matrix} T^{++}(2\omega) + G_{\mathrm{PSF}}(2\omega) \\ T^{+-}(2\omega) \end{matrix} \right\}, \quad (2)$$

where $\varepsilon(\vec{q})$ is the energy of an exciton with center-of-mass momentum $\vec{q}$, γ_2 is a phenomenological dephasing rate for the interband polarization, and C is a real, positive constant. The phase-space-filling term is $G_{\mathrm{PSF}}(\Omega) = -(4\pi/7)[(\hbar\Omega/2) - \varepsilon(\vec{0}) + i\gamma_2]a_0^2$ (a_0 is the 3D excitonic Bohr radius) and $T^{++}(\Omega)$ and $T^{+-}(\Omega)$, as discussed below, may be identified with the forward scattering amplitudes of two excitons with zero relative momentum and total center-of-mass frame energy $\hbar\Omega$. The $(2e, 2h)$ Hamiltonian is block diagonal in the basis of total electron spin [16]. Denoting the T's in the triplet (singlet) electron-spin channel by $T^{xx(+)}(T^{xx(-)})$, we have $T^{++} = T^{xx(+)}$, $T^{+-} = (T^{xx(+)} + T^{xx(-)})/2$. In each (electron spin) channel, T^{xx} is given by the equation

$$T^{xx(\lambda)}(\Omega + i\gamma_b) = W_{\vec{0},\vec{0}}^{xx(\lambda)*} + \sum_{\vec{q}\vec{q}'} W_{\vec{q},\vec{0}}^{xx(\lambda)*}[(\hbar\Omega - H^{xx(\lambda)} + i\gamma_b)^{-1}(1 - \lambda S)^{-1}]_{\vec{q},\vec{q}'} W_{\vec{q}',\vec{0}}^{xx(\lambda)}, \quad (3)$$

where $\lambda = \{+, -\}$ and the two-exciton Hamiltonian $H^{xx(\lambda)}$, in the exciton momentum basis, is

$$H_{\vec{q},\vec{q}'}^{xx(\lambda)} = 2\varepsilon(\vec{q})\delta_{\vec{q}\vec{q}'} + \sum_{\vec{k}}(1 - \lambda S)_{\vec{q},\vec{k}}^{-1} W_{\vec{k},\vec{q}'}^{xx(\lambda)}. \quad (4)$$

γ_b is an effective phenomenological decay rate of the coherent-biexciton amplitude, and $W_{\vec{q},\vec{q}'}^{xx(\lambda)}$ is the Coulomb matrix element between the initial exciton state with relative momentum $\vec{q}'$ and the final state $\vec{q}$. $W^{xx(\lambda)}$ includes both the direct and electron-exchange interactions, the latter being the dominant term at low momenta [6]. S is a matrix of overlap integrals between the nonorthonormal antisymmetrized two-exciton basis states. Further details of the two-exciton Hamiltonian can be found in [16–18].

In standard DCT treatments, the first and second terms on the right-hand side of Eq. (3) come from the Hartree-Fock and four-particle correlation contributions to the interband polarization, respectively. We would like to point out, however, that Eq. (3) can also be interpreted as the expression for the $\vec{q} = 0$ element of the off-energy-shell T matrix for two particles scattering off each other through the interaction $W^{xx(\lambda)}$, with two caveats: the presence of the matrix $(1 - \lambda S)^{-1}$ and the fact that $W^{xx(\lambda)}(\vec{q}, \vec{q}')$ is not Hermitian. Both complications can be traced to the nonorthogonality of the antisymmetrized two-exciton basis functions and the truncation of the expansion to $1s$, or, in other words, the fermionic composite nature of the exciton. Our numerical studies [19] show that, while omission of S and the anti-Hermitian part of $W^{xx(\lambda)}$ may change the result quantitatively, it does not affect the conclusions we draw below on the features of T^{xx} and on the comparisons with experiments. It is with these qualifications that we refer to T^{xx} as the exciton-exciton T matrix (or forward scattering amplitude).

The appearance of the exciton-exciton interaction $W_{\vec{q},\vec{q}'}^{xx(\lambda)}$ in the inverse propagator in Eq. (3) is equivalent to a nonperturbative, infinite-order dependence of the T matrix on the interaction W^{xx}. Thus, calculation of the T matrix requires knowledge of the true two-exciton scattering wave functions. Within the second Born approximation, the interaction term in the two-exciton Hamiltonian (4) is being neglected, so that, without the overlap matrix, the T matrix has the simple form

$$T^{xx(\lambda)}(\Omega + i\gamma_b) = W_{\vec{0},\vec{0}}^{xx(\lambda)*} + \sum_{\vec{q}} \frac{|W_{\vec{q},\vec{0}}^{xx(\lambda)}|^2}{\hbar\Omega - 2\varepsilon(\vec{q}) + i\gamma_b}. \quad (5)$$

This approximation corresponds to replacing the exact

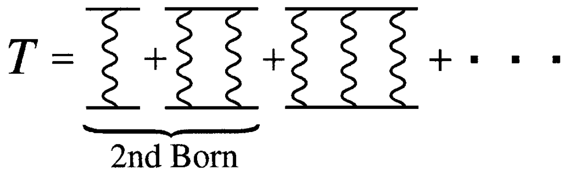

FIG. 1. Schematic visualization of the T matrix [Eq. (3)]. The squiggly lines represent the exciton-exciton interaction W^{xx}.

two-exciton wave function by a product of two plane waves. A schematic visualization of the T matrix and its second Born approximation is given in Fig. 1. One can see that, provided $\lim_{\vec{q}\to 0} W^{xx(\lambda)}_{\vec{q},\vec{0}} \neq 0$, the momentum sum in Eq. (5) develops a logarithmic singularity in its real part and a discontinuity in its imaginary part at $\hbar\Omega - 2\varepsilon(\vec{0}) = 0$ in the limit $\gamma_b \searrow 0$.

We have numerically evaluated T^{xx} by discretizing and diagonalizing H^{xx}, and constructing the resolvent in Eq. (3) via eigenfunction expansion [19]. In Fig. 2 we show, as an example, our calculated exciton-exciton T matrix for GaAs parameters (electron mass $m_e = 0.067$ in units of the electron mass in vacuum, hole mass $m_h = 0.1$, and background dielectric constant = 13) for the $++$ configuration. For clarity, the decay rate γ_b, which describes deviations from the ideal coherent $\chi^{(3)}$ regime, has been chosen to be small, $\gamma_b = 0.1$ meV (in high-quality quantum wells at low temperatures 0.1 meV seems to be a realistic value, cf. [20]). Figure 2 shows clearly the shortcomings of the second Born approximation. T^{+-} (not shown) carries a biexciton pole which modifies the low-energy continuum amplitude, worsening the errors in the 2nd Born T^{+-}.

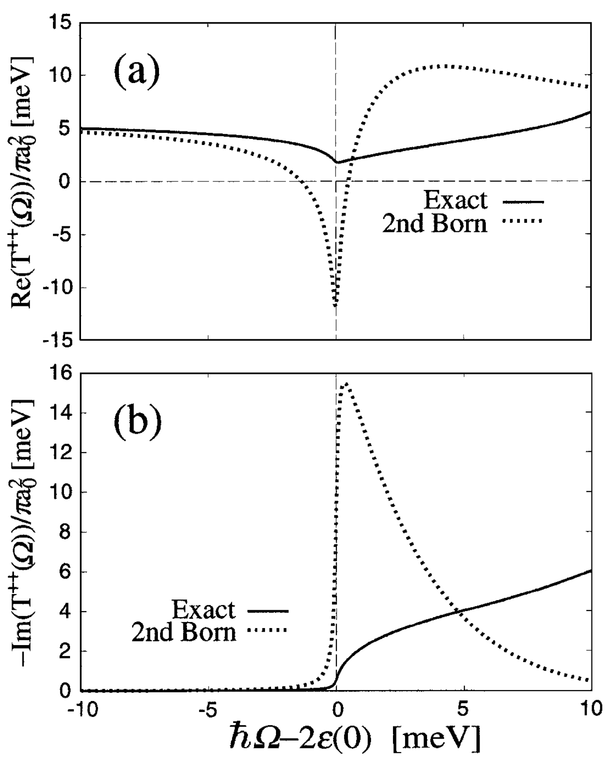

FIG. 2. Calculated exciton T matrix for GaAs quantum wells in the $(++)$ polarization configuration for $\gamma_b = 0.1$ meV. A small γ_b is used in this figure to highlight the developing divergence at zero energy. $\varepsilon(0)$ is the optical exciton frequency.

Microcavity FWM data.—For a quantum well in a microcavity, schematically shown in Fig. 3, we augment the above FWM theory with a standard transfer matrix treatment of light propagation through the dielectric layers (see, e.g., [21]). The quantum well is assumed to be very thin, so that its transfer matrix reduces to the one given as Eq. (29) in Ref. [21]. The transfer matrix solutions are done separately for the 1st-order and 3rd-order fields, with zero incoming fields on one (both) side(s) of the cavity for the 1st (3rd)-order fields. Since, in the experiment, the angle between pump and probe is very small, all light fields in the theory are assumed to come in at normal incidence. The solution of the 1st-order fields at the position of the quantum well is used as the source of the 3rd-order field which propagates in the FWM direction (see Fig. 3). The detectable FWM signal outside the cavity can then be written as $I_i^{\mathrm{FWM}}(\omega) \propto K(\omega)\,|P_i^{(3)}(\omega)|^2$ where $i = x, y, +$, or $-$ denotes the linear and circular polarization states of the signal, $P_i^{(3)}$ is evaluated with the appropriate choice of incident light-field polarization components, and $K(\omega)$ is a polarization-independent cavity factor that results from the transfer matrix solution [18]. The frequency dependence of $K(\omega)$ is similar to the linear reflectivity of the cavity, i.e., it exhibits peaks at the spectral positions of the cavity polariton modes (cf. Fig. 4). We adjust the width of these modes in order to account for finite pulse duration effects.

In order to address the question of whether a perturbative 2nd Born treatment of exciton-exciton scattering in two dimensions is justified, we compute the cavity FWM signal $I^{\mathrm{FWM}}(\omega)$ with material and cavity parameters corresponding to the experimental situation in Ref. [12]. We do so for both cases: (i) the T matrix evaluated in the 2nd BA and (ii) the full, nonperturbative solution for the T matrix. As one can see from Fig. 4, the full T-matrix solution gives excellent agreement with the experiment in all four polarization configurations. In contrast, the 2nd BA yields significant discrepancies,

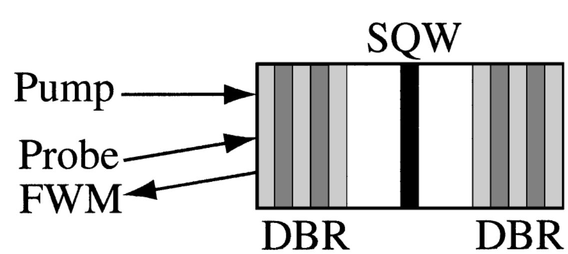

FIG. 3. Schematic of the microcavity four-wave mixing configuration (DBR = distributed Bragg reflector; SQW = single quantum well).

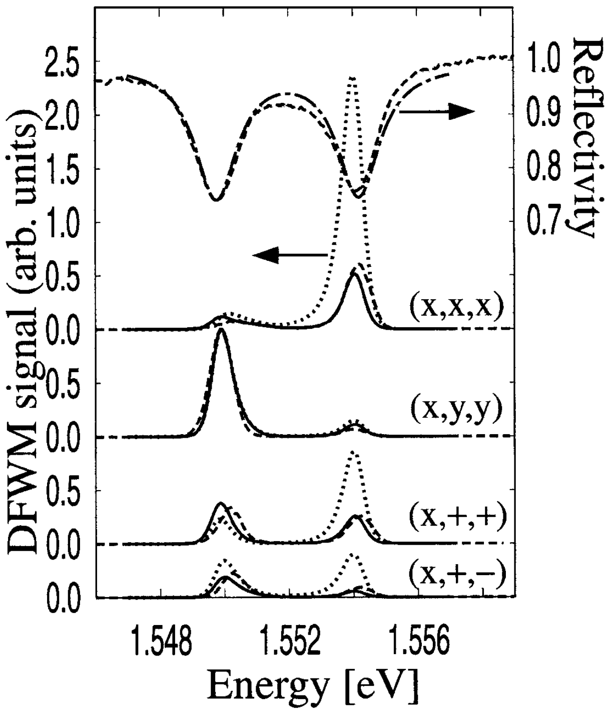

FIG. 4. Frequency-degenerate four-wave mixing signals from a semiconductor–quantum-well microcavity: calculated full T matrix solution (solid line), calculated signal in 2nd Born approximation (dotted line), experimental data taken from [12] (dashed line). The polarization configurations are indicated in the order (pump, probe, signal). Also shown is the linear reflectivity (upper panel: theory, dash-dotted line; experiment, dashed line). The cavity is tuned to the exciton resonance at 1.552 eV. In the calculation, the dephasing rates, $\gamma_2 = 0.75$ meV, $\gamma_b = 1.5$ meV, have been chosen in accordance with the experimental conditions in [12].

mainly at the upper cavity polariton peak. In this figure, we have normalized all theoretical data with the same normalization factor. This normalization factor was chosen such that the peak height of the T-matrix calculations at the lower peak in the (x, y, y) configuration coincides with the experimental peak height. Since the full T-matrix results agree so well with the experiment, we could have chosen any other peak to determine this factor without changing the basic appearance of Fig. 4. In particular, our conclusion—the 2nd Born approximation strongly overestimates the strength of the upper cavity-polariton FWM signal and hence the two-exciton continuum correlations—is independent of the way we choose the normalization factor. Our classical-light treatment neglects radiative corrections to the intermediate exciton states in Eq. (3) which, we believe, are unlikely to affect our conclusion since they do not change the available exciton density of states substantially. A more rigorous examination of this issue is under way. In summary, we have shown that microcavity FWM signals are sensitive probes of the exact nature of exciton-exciton correlations in quasi-two dimensions. The theory-experiment comparison presented above indicates the failure of the 2nd Born approximation, which we attribute to the fact that in *two* dimensions theoretical treatments of low-energy two-particle scattering have to be nonperturbative.

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