Coherent transient in photoluminescence of excitonic molecules in GaAs quantum wells

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Time-resolved photoluminescence (PL) of excitonic molecules in GaAs quantum wells (QW's) reveals an initial transient characterized by a finite rise and an extremely fast nonexponential decay [H. Wang, J. Shah, and T. C. Damen *et al.*, Solid State Commun. **98**, 807 (1996)]. The transient is attributed to coherent quantum evolution towards the molecule ground state of two optically correlated excitons, σ^+ and σ^-, which undergo the Coulombic attraction. In the present paper, we develop a theory of the transient PL and fit the experimental data. The radiative decay of a quasi-two-dimensional (2D) excitonic molecule is analyzed with the giant oscillator strength model adapted to QW's and with the bipolariton model. Within the 2D bipolariton model, the main ''hidden'' channel of the radiative decay of an excitonic molecule is the resonant dissociation into two outgoing interface (surface) polaritons rather than the observable decay into the bulk radiative modes. We conclude that quasi-2D molecules dissociate already within the initial transient of PL, while the following exponential decay of the molecule-mediated PL is due to the escape of secondary interface polaritons into the bulk modes. According to the 2D bipolariton model, the sequence ''two incoming photons→two virtual excitons→2D molecule→two outgoing interface polaritons'' is a completely coherent process. The fit of the time-resolved molecule-mediated PL provides us with numerical estimates of the exciton-photon coupling in GaAs QW's. [S0163-1829(97)08431-2]

I. INTRODUCTION

The quasi-2D excitonic molecules (m's) contribute to the various linear and nonlinear resonant optical processes in GaAs QW's. The m PL has been reported in Refs. 1–5, the m resonance has been detected in the time-resolved differential absorption,[6,7] and the m-mediated transient four-wave mixing (FWM) has been studied in Refs. 8–14.

Subpicosecond radiative dynamics in lower dimensional systems such as GaAs QW's is a subject of intense experimental and theoretical efforts. While the exciton (x) dynamics after resonant or nonresonant optical excitations has been extensively investigated,[15–17] very little information is available on the radiative dynamics of quasi-2D excitonic molecules in these structures. Recent PL measurements[18] with femtosecond resolution have shown a delayed rise of the x PL following a resonant excitation. The rise is determined by the initial exciton momentum relaxation, mainly due to the *density-dependent* $x(\sigma^{+(-)})-x(\sigma^{+(-)})$ Coulombic scattering.

A preliminary discussion of the radiative dynamics with 100 fs resolution of quasi-2D excitonic molecules in GaAs QW's has been reported in a previous paper.[4] Here, the observation of the coherent transient at the very beginning of the m-mediated PL is interpreted in terms of distructive quantum interference between the m ground state and x-x continuum. The *density-independent* temporal behavior of the initial transient stems from the quantum evolution towards the m bound state of two excitons, σ^+ and σ^-, optically correlated at time $t=0$ by the short pump pulse. The $x(\sigma^+)-x(\sigma^-)$ Coulombic attraction causes this coherent motion and gives rise to the inherently decaying beats between the m ground state and the x-x dispersive continuum. The importance of the wave vector dispersion of the x-x continuum has been emphasized in Ref. 19 for the m-mediated FWM.

In this paper we are mainly interested in the theoretical model and numerical simulations of the experimental results on the m-mediated transient PL. Our main result is a theoretical evidence that the resonant radiative dissociation of a quasi-2D molecule into QW polaritons is considerably more effective than the observable decay into the bulk radiative modes. In addition, we model a complete kinetics of the m-mediated PL and, in particular, estimate numerically the oscillator strength of the exciton-photon coupling in GaAs QW's. The rest of Introduction summarizes the experimental

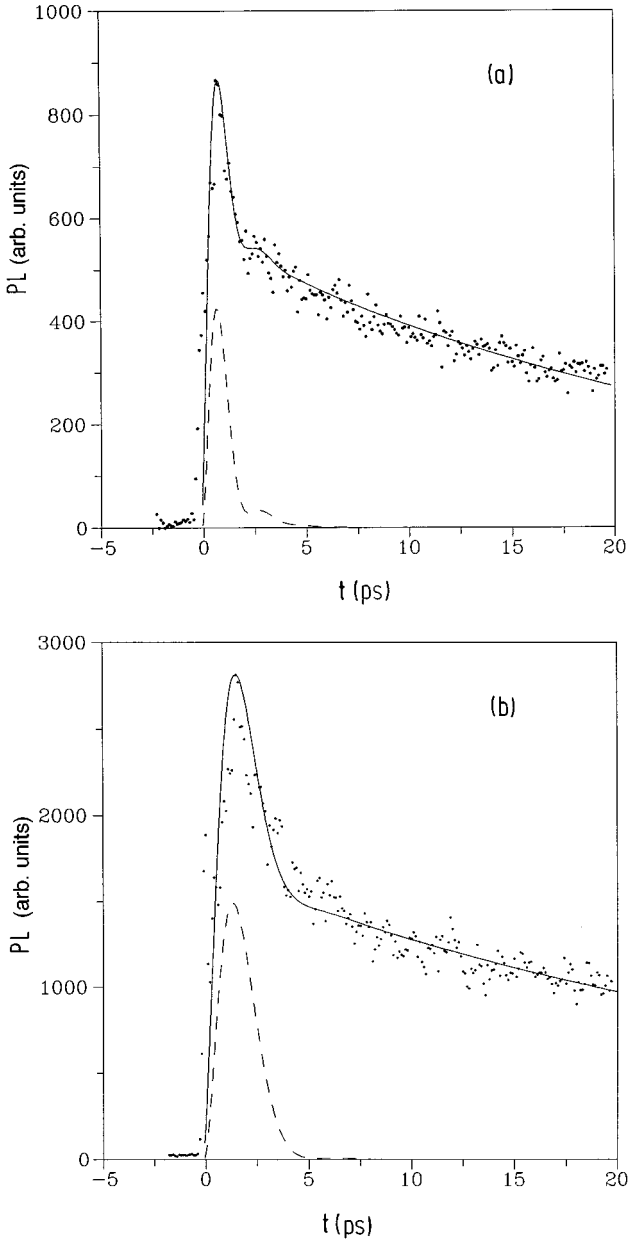

FIG. 1. Time-resolved cross-polarized PL from GaAs QW's samples no. 1 (a) and no. 3 (b). Points, experiment; solid line, fit within the 2D bipolariton model; dashed line contribution of excitonic molecules to the resonant photoluminescence.

data of Ref. 4. We also provide some additional arguments that the initial transient of the m-mediated PL is due to inherently decaying quantum beats between the m ground state and the x-x dispersive continuum.

Three high-quality samples have been used in our measurements and qualitatively the same behavior observed in all three samples. Sample no. 1 (no. 2) contains 15 periods of 130 Å (175 Å) GaAs QW and 150 Å $Al_{0.3}Ga_{0.7}As$ barrier and sample no. 3 contains four periods of 200 Å GaAs QW and 150 Å $Al_{0.3}Ga_{0.7}As$ barrier. The temporal evolution of the m-mediated cross linearly polarized PL is shown in Figs. 1(a) and 1(b) (points refer to the experimental data) for samples no. 1 and no. 3, respectively. The m-mediated PL reveals an initial transient characterized by a finite rise and an extremely fast nonexponential decay. A much slower exponential decay follows the initial transient. In the density range 10^9 to 10^{10} cm^{-2} the rise time of the initial transient is independent of the excitation level and of the temperature $T<30$ K (see Figs. 1 and 2 of Ref. 4).

The initial transient of the m-mediated PL cannot be explained within a standard picture of incoherent luminescence, which implies an *instanteneous* PL signal right after the optical excitation at $t\simeq 0$. We attribute the very first stage of the cross-polarized PL to a transient coherent quantum evolution towards the m ground state of two x's, σ^+ and σ^-, virtually excited at $t\simeq 0$ by the femtosecond pulse with duration $\tau_{\rm pulse}\ll\hbar/\epsilon_0^m$ (ϵ_0^m is the m ground-state binding energy). The optical pump induces coherently the real and virtual x's with fixed in-plane momentum $\mathbf{P}=\hbar\mathbf{k}_\parallel\ll\hbar/a_m$, due to momentum conservation along the QW. Here, $\mathbf{k}_\parallel$ is the in-plane component of the wave vector of the pump light and a_m is the quasi-2D m radius. These x's cannot emit photons (γ's) in the detection direction. On the other hand, such an *impact optical excitation* provides the well-defined initial condition $\Psi(\mathbf{p},t=0)=\delta(\mathbf{p})$ for the wave function of relative motion of two optically correlated x's.

In the absence of any dephasing, two σ^+ and σ^- excitons start to evolve coherently from the above initial condition due to the resonant $x(\sigma^+)-x(\sigma^-)$ attraction. This coherent quantum evolution given by the wave function of relative motion $\Psi(\mathbf{p},t)$ can be described by the time-dependent Schrödinger equation. Alternatively, one can expand the wave function:

$$\Psi(\mathbf{p},t)=\Psi_m^{(0)}(0)\Psi_m^{(0)}(\mathbf{p})e^{i\epsilon_0^m t}+\sum_{\mathbf{p_0}} c_{\mathbf{p_0}}\Psi_{\mathbf{p_0}}(\mathbf{p})e^{-i\epsilon(p)t}, \tag{1}$$

where $\Psi_m^{(0)}(\mathbf{p})$ is the ground-state m wave function and $\{\Psi_{\mathbf{p_0}}(\mathbf{p})\}$ is the x-x unbound eigenstates. We assume that a quasi-2D m in GaAs QW's has no excited states well-separated from the continuum. In further numerical evaluation of Eq. (1) the approximation of free-particle x-x continuum states is used, i.e., $\Psi_{\mathbf{p_0}}(\mathbf{p})=\delta(\mathbf{p}-\mathbf{p_0})$ and $\epsilon(p)=p^2/M_x$, where M_x is the x translational mass. The expansion coefficients $c_{\mathbf{p_0}}$ are then determined by the initial condition $\Psi(\mathbf{p},t=0)=\delta(\mathbf{p})$.

The function $|\Psi(\mathbf{p},t)|^2$, which describes the quantum coherent relative motion of two correlated x's, is plotted in Fig. 2 for times $t_n=n\pi\hbar/(2\epsilon_0^m)$ $(n=0,1,2,3,4)$. Here, we use the 2D deuteron wave function $\Psi_m^{(0)}(\mathbf{p})=2\sqrt{2\pi}a_m/(p^2a_m^2+1)^{3/2}$ for the m ground state [in real space $\Psi_m^{(0)}(\mathbf{r})=\sqrt{2/(\pi a_m^2)}\exp(-r/a_m)$]. According to Fig. 2, at $t=0$ the wave packet is concentrated at $\mathbf{p}=0$. Then, the wave packet isotropically spreads in momentum space up to momentum $p\sim\hbar/a_m$ and at $t=\pi\hbar/\epsilon_0^m$ starts to evolve back to the initial form. The wave packet approaches again its initial form at $t=2\pi\hbar/\epsilon_0^m$. Then, the motion repeates itself. The beats between the m ground state and the x-x continuum eventually damps out within a few periods due to complicated interferences involving simultaneously all modes $p\leqslant\hbar/a_m$. These inherently decaying oscillations differ from usual quantum beats in transient optical measurements which are due to the interference of the optical polarization field of different discrete transitions. Moreover, the transient m-mediated PL

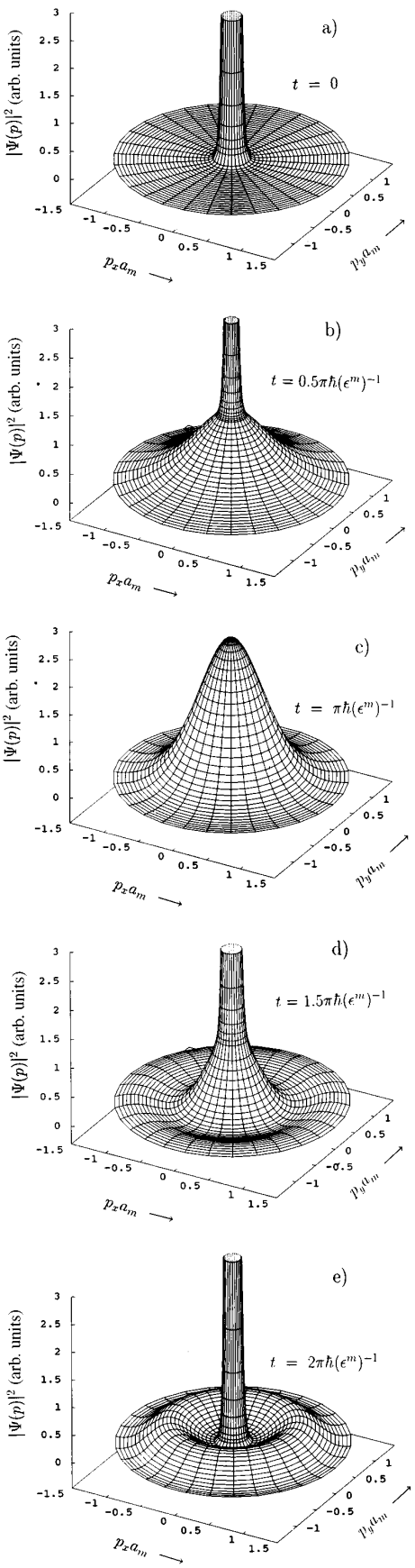

FIG. 2. Temporal evolution in momentum space of the m-mediated wave function $|\Psi(\mathbf{p},t)|^2$.

cannot be modeled within the few-level optical Bloch equations[11,12] where the x-x continuum is approximated by a discrete level.

The initial coherent stage of the m-mediated PL is governed by the time dependent wave function $\Psi[(\mathbf{k}_\parallel'-\mathbf{k}_\parallel)/2,t]$, where the in-plane wave vector $\mathbf{k}_\parallel'$ characterizes the direction of observation of the cross-polarized PL. The finite rise of the m-mediated PL is due to the coherent evolution of the reduced relative momentum from $\mathbf{p}=0$ to $\mathbf{p}=\pm(\mathbf{k}_\parallel'-\mathbf{k}_\parallel)/2$ caused by the $x(\sigma^+)-x(\sigma^-)$ Coulombic attraction. The coherent stage of the PL allows us to estimate the m binding energy ϵ_0^m by

$$\epsilon_0^m=\frac{\pi\hbar}{\tau(I_\perp^{\max})}, \tag{2}$$

where $\tau(I_\perp^{\max})$ is the rise time of the cross-polarized PL. Physically, Eq. (2) is consistent with the basic result of quantum mechanics: according to the time-energy uncertainty principle one cannot measure the m binding energy ϵ_0^m quickly than during $\Delta t\sim\hbar/\epsilon_0^m$. The finite rise time $\tau(I_\perp^{\max})$ of PL determines the duration of the coherent ''measurement'' of ϵ_0^m.

Within the above picture, one deals with Bloch coherence: right after an impact optical excitation the evolution of excitons follows the initial condition. On the other hand, the initial transient of the m-mediated PL is insensitive to photon statistics of the pump pulse. Basically, several important features distinguish the m transient from similar collisionless vibrational transients in polyatomic molecules:[20,21] (i) the m-mediated quantum beats occur for the *electronic* excitations (excitons) rather than for the considerably more heavy atoms; (ii) the $x(\sigma^+)-x(\sigma^-)$ quantum motion under the Coulombic attraction is described in momentum, rather than in real, space; (iii) the complicated interference in the m transient involves the x-x continuum rather than a quasi-equidistant discrete spectrum as for atomic oscillations.

According to the experimental data (see Figs. 1a,b and Fig. 2 of Ref. 4), only the first half-period of the m-mediated quantum beats is observed in the coherent initial transient. Therefore, in order to develop a complete kinetics of the cross-polarized PL one needs to analyze the corresponding dephasing and damping processes. Formally, the dephasing rate influences the off-diagonal terms of $|\Psi(\mathbf{p},t)|^2$ given by Eq. (1), while the damping rate is presented in the both diagonal and off-diagonal terms. In GaAs QW's, the x dephasing time is typically $\tau_c^x=1.3-6.0$ ps.[8–11,18] As will be shown below, the strongest damping process in the m-mediated PL, which even dominates over the dephasing, is the radiative dissociation of quasi-2D molecules into interface (surface) polaritons. The m radiative decay is determined by the x-photon coupling in QW's.

In Sec. II, the radiative decay of a quasi-2D m in GaAs QW's is examined theoretically. We discuss briefly the QW (interface) polaritons and bulk radiative modes. The giant oscillator strength model is adapted to the optical decay of a quasi-2D molecule into the bulk modes. Alternatively, a possible dissociation of m's into QW polaritons is analyzed. For this purpose, we generalized the recently developed exactly solvable bipolariton model of an excitonic molecule[22] in or-

der to include the both radiative and confined (interface) modes. Within the generalized 2D bipolariton model, it is shown that the dissociation of molecules into QW polaritons is much more effective than the m radiative decay into the bulk modes.

In Sec. III, we apply the 2D bipolariton model to describe the complete kinetics of the m-mediated PL from GaAs QW's. The model yields a dissociation of m's already during the initial transient of the cross-polarized PL. The fit of the experimental data allows us to estimate the oscillator strength of the x-γ coupling in the QW's. We also discuss a spatial anisotropy of the m-mediated PL.

II. OPTICAL DECAY OF A QUASI-2D EXCITONIC MOLECULE

In this section we analyze the radiative decay of a quasi-2D excitonic molecule both within the giant oscillator strength model adapted to QW's and with the 2D bipolariton model. It will be shown that the main ''hidden'' channel of the decay of m's in QW's is the dissociation into two outgoing QW polaritons rather than the observable radiative decay into bulk modes.

A. Surface polaritons and bulk radiative modes in GaAs quantum wells

The conventional polariton concept gives an adequate description of the exciton-photon system of a bulk direct-band-gap semiconductor, if the x-γ interaction is dominant. Due to the momentum conservation in the optical transition ''photon↔exciton,'' an incoming photon with momentum $\hbar\mathbf{p}$ can be many times resonantly reabsorbed and re-emitted by the x's with the same momentum. Within the polariton picture, this process is coherent and does not lead to ''true optical absorption.'' The dispersion law of bulk polaritons $c^2p^2/(\epsilon_0\omega^2)=1+\Omega_c^2/[\omega_x^2(p)-\omega^2]$ gives rise to the upper $[\omega=\omega^{pol}(p)=\omega^+(p)]$ and lower $[\omega=\omega^{pol}(p)=\omega^-(p)]$ dispersion branches. Here, $\omega_x(p)=\omega_t+\hbar p^2/2M_x$ is the x dispersion, $\hbar\omega_t$ is the energy of the transverse x, ϵ_0 is the background dielectric constant, and the polariton parameter Ω_c characterizes the exciton-photon coupling. The polariton states have been observed in bulk GaAs.[23] The polariton dispersion $\omega=\omega^{\pm}(p)$ is plotted in Fig. 3 for bulk GaAs (solid lines). Following the experimental data of Ref. 23, we use the longitudinal-transverse splitting $\hbar\omega_{lt}=0.08$ meV which gives $\hbar\Omega_c=\hbar\sqrt{2\omega_{lt}\omega_t}=15.6$ meV.

The polariton eigenstates are more complicated for quasi-2D QW's. In this case, the momentum conservation in the exciton-photon interaction holds only along a QW in the x-y plane. In the z direction (a growth direction) there is no translational invariance and, therefore, the photon momentum is not conserved in resonant absorption or emission. This simple argument allows us to classify two kinds of modes, bulk and confined ones.[24–26] Bulk or radiative modes, which are activated in conventional optical experiments, correspond to a plane-wave light field far outside ($z\to\pm\infty$) from the QW. These modes lie inside a photon cone, i.e., $p\leqslant\omega\sqrt{\epsilon_0}/c$. Surface or confined modes are associated with interface (surface) QW polaritons. The electromagnetic field of interface polaritons decays exponentially outside the QW,

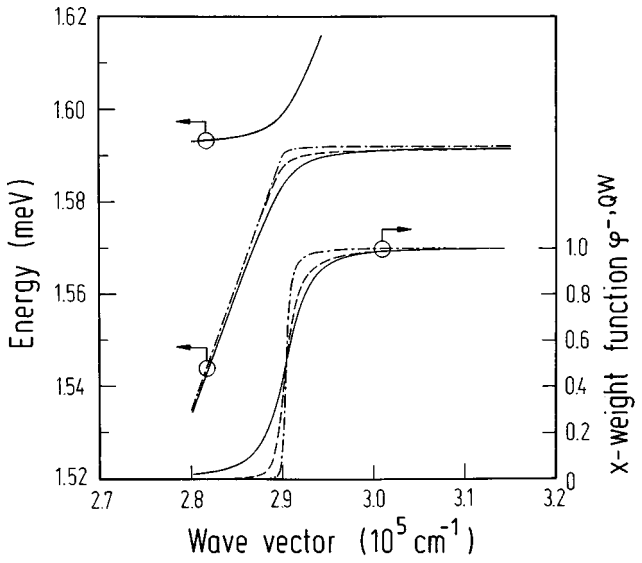

FIG. 3. The polariton dispersion in bulk GaAs (solid lines) and quasi-2D GaAs QW's (interface polaritons, $\hbar^2R_c=6.0\times10^{-2}$ eV^2 Å, dashed-dotted line, and $\hbar^2R_c=30.7\times10^{-2}$ eV^2 Å dashed line). The corresponding dependences $\varphi^{-,QW}=\varphi^{-,QW}(p)$ for bulk and interface (surface) polaritons. The following parameters of GaAs have been used: $\hbar\omega_t=1.592$ eV, $\hbar\Omega_c=15.6$ meV, $\epsilon_0=12.9$, and $M_x=0.44m_0$.

i.e., they are localized in the z direction. These modes cannot be excited straightforwardly by means of the external light, i.e., they are ''nonradiative.'' In other words, the 2D polariton picture, which needs the momentum conservation in the resonant exciton-photon coupling, holds only in the x-y plane of the QW. Roughly, with increasing confinement in z direction the upper dispersion branch of the bulk polaritons gives rise to radiative modes, while the lower one transforms into confined modes associated with the interface polaritons.

A complete classification of interface excitonic polaritons of a quasi-2D QW has been developed in Refs. 25 and 26. For a fixed polariton wave vector p_x along the x axis, there are X-, Y-, and Z-mode interface polaritons in accordance with polarization of the light field. The X (Y,Z) mode refers to a polarization parallel to the x (y,z) axis. We concentrate on the Y-mode transverse interface polaritons with in-plane wave vector $\mathbf{p}$ and polarization parallel to the QW. The 2D bipolariton model deals with these polaritons. The dispersion equation for Y-mode polaritons is given by[25]

$$\frac{c^2p^2}{\epsilon_0}=\omega^2+\frac{\omega^2R_c\sqrt{p^2-\omega^2\epsilon_0/c^2}}{\omega_t^2+\omega_tp^2/M_x-\omega^2}, \tag{3}$$

where the parameter R_c is proportional to the oscillator strength per unit area of the QW and refers to the quasi-2D exciton-photon coupling. The 2D QW polaritons are sensitive to the design of a concrete semiconductor microstructure. Equation (3) implies a symmetric GaAs QW sandwiched in between two identical GaAlAs barriers. The thickness d of the well corresponds to the long wavelength limit, i.e., satisfies the condition $pd\ll1$.

For narrow-width QW's ($d\to0$), one can estimate the x-photon coupling parameter in Eq. (3) as $R_c=\Omega_c^2|\varphi^{(2D)}(0)/\varphi^{(3D)}(0)|^2=16a_x^{(3D)}\omega_{lt}\omega_t$. Here, $\varphi^{(2D)}(r)$ and

$\varphi^{(3D)}(r)$ are the real-space wave functions of the ground-state 2D and 3D x's, respectively, the corresponding 2D and 3D x Bohr radii satisfy the condition $a_x^{(2D)}=a_x^{(3D)}/2$, and the polariton parameters Ω_c and ω_{lt} refer to bulk material. For GaAs QW's, we estimate the upper limit of the coupling parameter as $\hbar^2 R_c \simeq 30.7\times 10^{-2}$ eV^2 Å. The Y-mode polariton dispersions $\omega=\omega^{QW}(p)$ given by Eq. (3) are plotted in Fig. 3 for $\hbar^2 R_c=6.0\times 10^{-2}$ eV^2 Å (dashed-dotted line) and $\hbar^2 R_c=30.7\times 10^{-2}$ eV^2 Å (dashed line). Figure 3 traces an evolution of the lower polariton branch $\omega=\omega^{-}(p)$ towards the interface polariton dispersions.

The Y-mode dispersion shown in Fig. 3 reflects the following general tendency. The quantum confinement in QW's enhances the exciton-photon coupling in comparison with that in bulk material. The coupling parameter R_c is responsible for this effect. On the other hand, from comparison of the dispersions of 3D and QW polaritons, one can introduce an effective polariton parameter $[\Omega_c(\mathbf{p},\omega)]^2 = R_c\sqrt{p^2-\omega^2\epsilon_0/c^2}$. This parameter provides the fast decay of the polariton effects with a decrease of $\omega=\omega^{QW}(p)$ [$\omega^{QW}(p)\rightarrow cp/\sqrt{\epsilon_0}$ for $p\rightarrow 0$]. The photon factor $\sqrt{p^2-\omega^2\epsilon_0/c^2}$ is responsible for such a behavior. The 2D polaritons have been observed in GaAs QW's.[27]

One can still apply Eq. (3) to describe the radiative modes located within the photon cone given by $p_0(\omega)=\omega\sqrt{\epsilon_0}/c$. In this case, $\sqrt{p^2-\omega^2\epsilon_0/c^2}=i\sqrt{p_0^2-p^2}$ and the frequency ω can be approximated by $\omega_t-i(\Gamma_{\mathbf{p}}^x/2)$, where $\Gamma_{\mathbf{p}}^x$ is the radiative decay rate of the quasi-2D exciton with momentum $\mathbf{p}$. Formally, $\Gamma_{\mathbf{p}}^x/2$ is the imaginary part of the complex frequency $\omega=\omega(p)$ which is the solution of Eq. (3) for real $p\leqslant p_0$. In the resonant approximation one gets

$$\Gamma_{\mathbf{p}}^x=\frac{\epsilon_0}{c^2}\,R_c\,\frac{\omega_t}{\sqrt{p_0^2-p^2}}=\Gamma_0^x\,\frac{p_0}{\sqrt{p_0^2-p^2}}, \tag{4}$$

where $\Gamma_0^x=(\sqrt{\epsilon_0}/c)R_c$ is the radiative width of the exciton with in-plane momentum $p=0$. Equation (4) is consistent with the results of Refs. 26. However, the main advantage of our approach is an explicit inclusion of the same coupling parameter R_c for a description of the both conjugated, confined and bulk, modes. The parameter R_c can be written in terms of the oscillator strength f_{xy} per unit area of the QW (the subscript xy refers to the light polarization parallel to the QW) by

$$R_c=\pi\,\frac{e^2}{\epsilon_0 m_0}\,f_{xy}, \tag{5}$$

where m_0 is the free electron mass. The extension of the QW polariton dispersion (3) on the bulk modes, i.e., Eq. (4), corresponds to the s polarization of the incident or emitted light.

B. Giant oscillator strength model for excitonic molecules in quantum wells

Usually, one separates the x-γ polariton coupling and the $x(\sigma^+)-x(\sigma^-)$ Coulombic attraction. First, the ground state of an excitonic molecule is evaluated variationally within the underlying electron-hole picture. This procedure yields the ground-state eigenfunction $\Psi_m^{(0)}(\mathbf{r})$ in the x representation. Then, the optical properties of the molecule are treated within the giant oscillator strength model.

The giant oscillator strength model applied for the optical creation of the molecule, real or virtual, operates with the following two-step elementary scheme: ''two incoming photons→exciton+photon→excitonic molecule''.[28,29] This two-photon process is effective due to (i) the nearly resonant intermediate x level, and (ii) the spatial extension of the m wave function. Within this scenario, the PL of m's deals with an optical conversion ''$m\rightarrow\gamma+$ recoil x.'' The 2D giant oscillator strength model formulates the m-mediated optics in terms of coupling of molecules with the radiative modes.

In order to adapt the giant oscillator strength model for excitonic molecules in QW's, we estimate the spontaneous optical decay of a quasi-2D m into the radiative modes as

$$\Gamma_{\text{bulk}}^m=2\int_0^{k_{\text{opt}}}|\Psi_m^{(0)}(2p)|^2\left[\Gamma_0^x\,\frac{k_{\text{opt}}}{\sqrt{k_{\text{opt}}^2-p^2}}\right]\frac{pdp}{2\pi}$$
$$=8(a_m k_{\text{opt}})^2\Gamma_0^x, \tag{6}$$

where $\Psi_m^{(0)}(p)$ is the deuteron wave function of the 2D m ground state and $k_{\text{opt}}=\omega_t\sqrt{\epsilon_0}/c$ ($k_{\text{opt}}\simeq 2.9\times 10^5$ cm^{-1} for GaAs) is the resonant wave vector of the crossover between the unperturbed dispersions of free photons and 2D excitons. Physically, the radiative width Γ_{bulk}^m is given by a product of two probabilities in the integrand of Eq. (6). The first one, $|\Psi_m^{(0)}(2p)|^2$, is the probability of two bound x's with the reduced relative momentum $2p$ to be in the radiative zone $p\leqslant k_{\text{opt}}$, i.e., within the photon cone. The second one, $\Gamma_{\mathbf{p}}^x$ (the term in the square brackets), is the probability per unit time that an exciton with momentum $p\leqslant k_{\text{opt}}$ emits a photon into the radiative modes. The numerical prefactor 2 in Eq. (6) is because the both x's equally contribute to the optical decay of the molecule.

According to the right hand side (r.h.s.) of Eq. (6), the 2D giant oscillator strength model contains a natural enhancement factor $a_m^2 \gg a_x^2$. In other words, due to the spatially extended m wave function $\Psi_m^{(0)}$, the x with in-plane momentum $\mathbf{p}$ acts like an ''antenna'' that collects (escapes) the second x with momentum $\mathbf{p}'$ from the whole coherent m area $\sim a_m^2$.

For $a_m=130$ Å ($\epsilon_0^m=2$ meV), we estimate from Eq. (6) $\Gamma_{\text{bulk}}^m\simeq 1.2\Gamma_0^x$. As a result, $\tau_{\text{bulk}}^m=\hbar/\Gamma_{\text{bulk}}^m\sim 20$ ps, i.e., the giant oscillator strength model yields approximately the same lifetime of a quasi-2D molecule as the radiative lifetime of the constituent x's [$\hbar/\Gamma_0^x\sim 25$ ps (Ref. 26)]. This result contradicts the conclusion of Ref. 30, where the giant oscillator strength model has been interpreted as misleading for quasi-2D excitonic molecules in QW's.

Within the 2D giant oscillator strength model one can indeed reproduce the m-mediated PL.[4] The resulting theoretical fit is shown by the dashed line in Fig. 2 of Ref. 4. Here, for sample no. 1 we have taken the coherence lifetime $\tau_c^m=1.3$ ps, the m radiative lifetime $\tau_{\text{bulk}}^m=27.4$ ps, and the m binding energy $\epsilon_0^m=1.6$ meV. In this approach, the initial transient of the m-mediated PL dies away due to the loss of the initial coherence and the following exponential decay is attributed to the observable decay of ground-state m's into the radiative modes.

C. Bipolariton model of quasi-2D excitonic molecules

An exactly solvable bipolariton model of a molecule has been developed recently[22] to include the exciton-photon coupling in an equal manner with the x-x attraction. The x-γ interaction influences the m wave function $\Psi_m^{(0)}(p)$ unperturbed by the polariton effects. Even in the low-excitation limit, both the $x(\sigma^{\pm})$ - photon ($\sigma^{\pm}$) and $x(\sigma^{+})-x(\sigma^{-})$ interactions have to be treated simultaneously and beyond a standard low-order perturbation theory. Within the bipolariton concept, the true components of an excitonic molecule in direct-band-gap semiconductors are two polaritons quasibound through the resonant Coulombic attraction, rather than two excitons. The solution of the bipolariton wave equation describes the m resonant decay into two outgoing polaritons as a continuous quantum evolution of the m internal state rather than as a discrete act of the optical conversion ''$m \rightarrow x+\gamma$.'' The importance of the bipolariton concept has been already recognized in Ref. 31 for molecules in bulk semiconductors.

Within the 2D bipolariton model, both the confined and bulk modes contribute to the internal state of a molecule. The resonant $x(\sigma^{+})-x(\sigma^{-})$ Coulombic attraction couples these modes in the relative motion of two quasibound x's. In other words, an ''umklapp'' of the x between surface and radiative modes is possible. However, the main contribution to the m state is due to the surface modes, because the radiative modes are located at $p \leqslant k_{\rm opt}$, while the relative motion of two constituent x's involves the momenta $p \sim a_m^{-1} \gg k_{\rm opt}$. Therefore, a quasi-2D molecule in QW's exists mainly as two quasibound QW polaritons and, consequently, radiatively dissociates primarily into two outgoing interface polaritons. This important feature is completely ignored in the conventional approaches to m-mediated optics of GaAs QW's. The 2D bipolariton model naturally emphasizes the role of the confined modes.

The influence of the polariton effects on the m ground state increases drastically with a reduction of the dimensionality from 3D to 2D. This is because the fraction of the phase space where the polariton effects dominate over the Coulombic $x(\sigma^{+})-x(\sigma^{-})$ attraction is roughly of the order of $(k_{\rm opt}a_m)^D$. One gets $(k_{\rm opt}a_m)^2 \sim 0.1$ for GaAs QW's. In Ref. 22, the exactly solvable 2D bipolariton model has been developed for the deuteron x-x attraction potential $W_{12}(p)$. This theory, which treats only the confined modes in a 2D bipolariton, yields $\tau_{QW}^m \simeq 0.5-3.0$ ps for the m spontaneous radiative dissociation into two outgoing interface polaritons (see the inset of Fig. 4). Therefore, τ_{QW}^m is indeed considerably shorter than $\tau_{\rm bulk}^m$ estimated within the 2D giant oscillator strength model. Here, we develop a 2D bipolariton theory which treats the both confined and bulk modes. This allows us to make a self-consistent comparison of the two decay mechanisms. Apart from a generalization of the exactly solvable 2D bipolariton model to include the radiative modes, we find a bipolariton solution valid for an arbitrary short-range potential W_{12}.

In order to describe a molecule as a (quasi) bound state of two excitons (polaritons) one can apply the standard diagram technique. Namely, the poles of the vertex function Γ, which refers to the $x(\sigma^{+})-x(\sigma^{-})$ Coulombic attraction, provide the m energies.

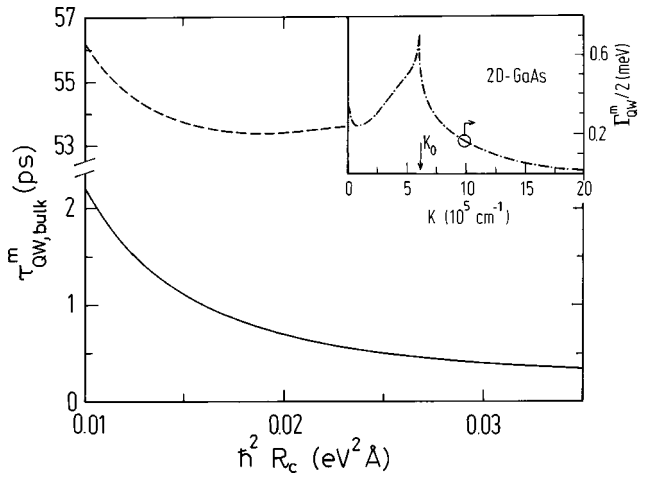

FIG. 4. The radiative lifetime $\tau_{QW}^m(K \simeq 0)$ (solid line) and $\tau_{\rm bulk}^m(K \simeq 0)$ (dashed line) versus the exciton-photon coupling parameter R_c. The m binding energy $\epsilon_0^m = 2$ meV. Inset: the radiative half-width $\Gamma_{QW}^m(\mathbf{K})/2$ due to the dissociation of a quasi-2D molecule into two interface polaritons in a narrow-width GaAs QW with $\hbar^2 R_c = 30.7 \times 10^{-2}$ eV^2 Å. The vertical arrow indicates the position K_0 of the van Hove singularity in the joint density of QW polariton states.

In the absence of the polariton effects, the Bethe-Salpeter equation for a quasi-2D molecule is given by

$$\Gamma_0(\mathbf{p},\mathbf{p}';K) = W_{12}(\mathbf{p}-\mathbf{p}') + \sum_{\mathbf{p}''} W_{12}(\mathbf{p}-\mathbf{p}'') \times \widetilde{G}_0(\mathbf{p}'',K)\Gamma_0(\mathbf{p}'',\mathbf{p}';K), \tag{7}$$

where $p=\{\mathbf{p},\omega\}$ and $p'=\{\mathbf{p}',\omega'\}$ are the reduced in-plane relative momenta and frequencies of two x's before and after an interaction event, $K=\{\mathbf{K},\Omega\}$ is the conserved in-plane total momentum and frequency. The two-x Green function $\widetilde{G}_0(\mathbf{p},K)$ is determined by $\widetilde{G}_0(\mathbf{p},K) = -(i/2\pi)\int d\omega G^{(0)}(-p+K/2)G^{(0)}(p+K/2) = [\Omega - \omega_x(-\mathbf{p}+\mathbf{K}/2)+\omega_x(\mathbf{p}+\mathbf{K}/2)]^{-1}$, where $G^{(0)}(p) = 1/[\omega - \omega_x(\mathbf{p}) + i\delta]$ is the free-x propagator. In a spectral vicinity of the ground-state m resonance, i.e., for $\Omega \simeq \Omega_{\mathbf{K}}^m$, one approximates

$$\widetilde{G}_0(\mathbf{p},K)\widetilde{G}_0(\mathbf{p}',-K)\Gamma_0(\mathbf{p},\mathbf{p}';K) = \frac{\Psi_m^{(0)}(\mathbf{p})\Psi_m^{(0)*}(\mathbf{p}')}{\Omega - \Omega_{\mathbf{K}}^m + i\delta}. \tag{8}$$

With this basic property of the vertex function Γ_0, the homogeneous part of the Bethe-Salpeter Eq. (7) reduces to

$$\widetilde{G}_0^{-1}(\mathbf{p},\mathbf{K},\Omega_{\mathbf{K}}^m)\Psi_m^{(0)}(\mathbf{p}) - \sum_{\mathbf{p}'} W_{12}(\mathbf{p}-\mathbf{p}')\Psi_m^{(0)}(\mathbf{p}') = 0. \tag{9}$$

From Eq. (9) one finally gets the standard wave equation for an excitonic molecule unperturbed by the polariton effects:

$$[\omega_x(\mathbf{p}+\mathbf{K}/2)+\omega_x(-\mathbf{p}+\mathbf{K}/2)]\Psi_m^{(0)}(\mathbf{p})$$

$$+\sum_{\mathbf{p}'} W_{12}(\mathbf{p}-\mathbf{p}')\Psi_m^{(0)}(\mathbf{p}')=\Omega_{\mathbf{K}}^m\Psi_m^{(0)}(\mathbf{p}). \quad (10)$$

Due to the quadratic x dispersion, the center-of-mass motion splits off and the wave function $\Psi_m^{(0)}(\mathbf{p})$ of the relative motion of two constituent x's as well as the corresponding m binding energy $\epsilon_0^m=2\omega_t+K^2/2M_m-\Omega_{\mathbf{K}}^m$ are independent of the translational in-plane momentum $\hbar\mathbf{K}$ of the molecule.

In the bipolariton model, we include the polariton effects in the Bethe-Salpeter equation:

$$\Gamma(\mathbf{p},\mathbf{p}';K)=W_{12}(\mathbf{p}-\mathbf{p}')+\sum_{\mathbf{p}''} W_{12}(\mathbf{p}-\mathbf{p}'')\times\widetilde{G}(\mathbf{p}'',K)\Gamma(\mathbf{p}'',\mathbf{p}';K), \quad (11)$$

where the two-x Green function $\widetilde{G}(\mathbf{p},K)=-i/(2\pi)\int d\omega G(-p+K/2)G(p+K/2)$ is now the frequency convolution of two x propagators dressed by the light field. In the 2D bipolariton model, the photon-mediated x Green function is given by

$$G(\mathbf{p},\omega)=G^{(0)}(\mathbf{p},\omega)+\tfrac{1}{4}[\Omega_c^{\mathrm{eff}}(\mathbf{p},\omega)]^2G^{(0)}(\mathbf{p},\omega)\times D^{(0)}(\mathbf{p},\omega)G(\mathbf{p},\omega)=\frac{\varphi^{QW}[\mathbf{p},\omega^{QW}(\mathbf{p})]}{\omega-\omega^{QW}(\mathbf{p})+i\delta}+\frac{\varphi^{\mathrm{bulk}}[\mathbf{p},\omega^{\mathrm{bulk}}(\mathbf{p})]}{\omega-\omega^{\mathrm{bulk}}(\mathbf{p})}, \quad (12)$$

where $D^{(0)}(\mathbf{p},\omega)=1/(\omega-cp/\sqrt{\epsilon_0}+i\delta)$ is the free-particle photon propagator in the resonant approximation and

$$\varphi^{QW}[\mathbf{p},\omega^{QW}(\mathbf{p})]=\frac{R_c(p^2-\omega^2\epsilon_0/c^2)^{1/2}}{R_c(p^2-\omega^2\epsilon_0/c^2)^{1/2}+4[\omega_x(\mathbf{p})-\omega^{QW}(\mathbf{p})]^2},$$

$$\varphi^{\mathrm{bulk}}[\mathbf{p},\omega^{\mathrm{bulk}}(\mathbf{p})]=1-\varphi^{QW}(\mathbf{p},\omega^{QW}(\mathbf{p})). \quad (13)$$

Here, $\omega^{QW/\mathrm{bulk}}(\mathbf{p})$ are the dispersions of the interface polaritons and the x radiative states, respectively. The last dispersion is approximated by $\omega^{\mathrm{bulk}}(\mathbf{p})=\omega_t-i(\Gamma_{\mathbf{p}}^x/2)$ for $p\leqslant k_{\mathrm{opt}}$. The initial dispersion Eq. (3) is cubic with respect to ω. However, only the two branches of its solution, i.e., $\omega^{QW/\mathrm{bulk}}(\mathbf{p})$, have a physical meaning (the third branch corresponds to an amplified wave).

The functions $\varphi^{QW/\mathrm{bulk}}$ characterize the distribution of the x state between the confined and bulk modes. The last relationship of Eqs. (13) is a ''sum rule'' for the photon–quasi-2D exciton transition. The x-weight functions $\varphi^{QW}[\mathbf{p},\omega^{QW}(\mathbf{p})]$ of interface polaritons and $\varphi^{-}[\mathbf{p},\omega^{-}(\mathbf{p})]$ of the lower dispersion branch of bulk polaritons in GaAs are plotted in Fig. 3.

Using Eq. (12) one derives

$$\widetilde{G}(\mathbf{p},K)=\sum_{\alpha,\alpha'}\frac{\varphi^{\alpha}[-\mathbf{p}+\mathbf{K}/2,\omega^{\alpha}(-\mathbf{p}+\mathbf{K}/2)]\varphi^{\alpha'}[\mathbf{p}+\mathbf{K}/2,\omega^{\alpha'}(\mathbf{p}+\mathbf{K}/2)]}{\Omega-\omega^{\alpha}(-\mathbf{p}+\mathbf{K}/2)-\omega^{\alpha'}(\mathbf{p}+\mathbf{K}/2)+i\delta}, \quad (14)$$

where $\alpha,\alpha'=QW/\mathrm{bulk}$. The x-γ resonant coupling modifies the m ground-state energy $\widetilde{\Omega}_{\mathbf{K}}^m=\Omega_{\mathbf{K}}^m+\Delta^m(\mathbf{K})-i\Gamma^m(\mathbf{K})/2$. Here, $\Delta^m(\mathbf{K})$ is the radiative renormalization of the m energy, i.e., the m Lamb shift. Within the bipolariton model, the 2D molecule is metastable due to the resonant interaction with the electromagnetic field. The both radiative and confined modes contribute to total $\Gamma^m(\mathbf{K})$. Similarly Eq. (8), one has in the resonant approximation of the m (bipolariton) ground state:

$$\widetilde{G}(\mathbf{p},K)\widetilde{G}(\mathbf{p}',-K)\Gamma(\mathbf{p},\mathbf{p}';K)=\frac{\widetilde{\Psi}(\mathbf{p},\mathbf{K})\widetilde{\Psi}^*(\mathbf{p}',\mathbf{K})}{\Omega-\widetilde{\Omega}_{\mathbf{K}}^m}, \quad (15)$$

where $\widetilde{\Psi}(\mathbf{p},\mathbf{K})$ is the bipolariton wave function. From Eqs. (11) and (15) we find the 2D bipolariton wave equation

$$\widetilde{G}^{-1}(\mathbf{p},\mathbf{K},\widetilde{\Omega}_{\mathbf{K}}^m)\widetilde{\Psi}(\mathbf{p},\mathbf{K})-\sum_{\mathbf{p}'} W_{12}(\mathbf{p}-\mathbf{p}')\widetilde{\Psi}(\mathbf{p}',\mathbf{K})=0, \quad (16)$$

where $\widetilde{G}(\mathbf{p},\mathbf{K},\widetilde{\Omega}_{\mathbf{K}}^m)$ given by Eq. (14) includes the bulk and radiative modes.

The 2D exactly solvable model has been developed in Ref. 22 for an interface (surface) bipolariton, i.e., *only the confined modes have been treated*. Here, we adapt this method to include the bulk modes. The bipolariton wave function can be written as $\widetilde{\Psi}(\mathbf{p},\mathbf{K})=\Psi_m^{(0)}(\mathbf{p})+\delta\Psi(\mathbf{p},\mathbf{K})$, where $\Psi_m^{(0)}(\mathbf{p})$ is a known solution of the standard m wave Eq. (10), while $\delta\Psi(\mathbf{p},\mathbf{K})$ is large only in the optical range, i.e., for $p\simeq k_{\mathrm{opt}}\ll a_m^{-1}$. With this substitution, Eq. (16) reduces to a Fredholm integral equation with separable kernel for $\delta\Psi(\mathbf{p},\mathbf{K})$. The solution of this equation yields

$$\widetilde{\Psi}(\mathbf{p},\mathbf{K})=\widetilde{G}(\mathbf{p},\mathbf{K},\widetilde{\Omega}_{\mathbf{K}}^m)[\Phi_m^{(0)}(\mathbf{p})+\widetilde{C}_{\mathbf{K}}W_{12}(\mathbf{p})], \quad (17)$$

where $\widetilde{C}_{\mathbf{K}}=\widetilde{A}_{\mathbf{K}}/(1+\widetilde{B}_{\mathbf{K}})$ and

$$\widetilde{A}_{\mathbf{K}}=\sum_{\mathbf{p}}[\widetilde{G}(\mathbf{p},\mathbf{K},\widetilde{\Omega}_{\mathbf{K}}^m)-\widetilde{G}_0(\mathbf{p},\mathbf{K},\Omega_{\mathbf{K}}^m)]\Phi_m^{(0)}(\mathbf{p}), \quad (18)$$

$$\widetilde{B}_{\mathbf{K}}=-\sum_{\mathbf{p}} W_{12}(\mathbf{p})\widetilde{G}(\mathbf{p},\mathbf{K},\widetilde{\Omega}_{\mathbf{K}}^m).$$

In Eqs. (17) and (18), $\widetilde{G}_0^{-1}(\mathbf{p},\mathbf{K},\Omega_{\mathbf{K}}^m)=-\epsilon_0^m-p^2/M_x$ and $\Phi_m^{(0)}(\mathbf{p})=\widetilde{G}_0^{-1}(\mathbf{p},\mathbf{K},\Omega_{\mathbf{K}}^m)\Psi_m^{(0)}(\mathbf{p})$.

For model potentials, which satisfy the condition $W_{12}(\mathbf{p})=\beta\Psi_m^{(0)}(\mathbf{p})$ with $\beta=\text{const}<0$, the bipolariton energy is determined by the equation

$$\widetilde{\Omega}_{\mathbf{K}}^m=\Omega_{\mathbf{K}}^m-\beta\widetilde{C}_{\mathbf{K}}(\widetilde{\Omega}_{\mathbf{K}}^m). \tag{19}$$

In this case $\delta\Psi(\mathbf{p},\mathbf{K})\to 0$, $\widetilde{\Psi}(\mathbf{p},\mathbf{K})\to\Psi_m^{(0)}(\mathbf{p})$ for $p\gg k_{\text{opt}}$ and only the poles of $\widetilde{G}(\mathbf{p},\mathbf{K},\widetilde{\Omega}_{\mathbf{K}}^m)$ contribute to $\delta\Psi(\mathbf{p},\mathbf{K})$. These poles are determined by the energy-momentum conservation $\widetilde{\Omega}_{\mathbf{K}}^m-\omega^{\alpha}(-\mathbf{p}+\mathbf{K}/2)-\omega^{\alpha'}(\mathbf{p}+\mathbf{K}/2)=0$ $(\alpha,\alpha'=QW,$ bulk) in the optical decay of the quasi-2D molecule. The condition (19) provides that the poles of $\Psi_m^{(0)}(\mathbf{p})$ at $p=\pm i/a_m$ do not contribute to $\delta\Psi(\mathbf{p},\mathbf{K})$. Equation (19) is transcendent with respect to the bipolariton energy $\widetilde{\Omega}_{\mathbf{K}}^m$. However, if the radiative corrections are small compared with the unperturbed m binding energy $(\Delta^m,\Gamma^m\ll\epsilon_0^m)$, one can use the iteration procedure and put $\widetilde{\Omega}_{\mathbf{K}}^m=\Omega_{\mathbf{K}}^m-\beta\widetilde{C}_{\mathbf{K}}(\Omega_{\mathbf{K}}^m)$.

The condition $W_{12}(\mathbf{p})\propto\Psi(\mathbf{p})$ indeed holds, e.g., for some particular realizations of the 2D deuteron $W_{12}(p)=W(0)/[(pa_m)^2+1]^{3/2}$ and Gauss $W_{12}(p)=W(0)\exp\times[-(pa_m)^2/4]$ potentials. Namely, it is valid for the deuteron potential with $W(0)=-27\pi/2$ (Fourier transform to real space is $W_{12}(r)=(-27/4)\exp(-r/a_m)$, where $r=\sqrt{x^2+y^2}$) and for the Gauss potential with $W(0)=-9\pi$ [in real space $W_{12}(r)=-9\exp(-r^2/a_m^2)$]. Here, the potential W_{12} is given in units of the Rydberg $M_x e^4/(2\hbar^2\epsilon_0)$.

The solution, Eqs. (17)–(19), implies a ''soft'' potential W_{12} which changes in momentum (real) space in the same scale as the wave function $\Psi_m^{(0)}$. Here, we construct an approximate solution of the 2D bipolariton model for a short-range ''contact'' potential W_{12}. In real space such a potential can be modeled by $W_{12}(\mathbf{r})\propto\delta(\mathbf{r})$, while in momentum space $W_{12}(\mathbf{p})$ can be approximated by $W_{12}(0)$ within the scale $\sim 1/a_m$ of the change of $\Psi_m^{(0)}(\mathbf{p})$. In this case, the second term in square brackets on the r.h.s. of Eq. (17) can be neglected, i.e., $\widetilde{\Psi}(\mathbf{p},\mathbf{K})=\widetilde{G}(\mathbf{p},\mathbf{K},\widetilde{\Omega}_{\mathbf{K}}^m)\Phi_m^{(0)}(\mathbf{p})$. Therefore, from Eqs. (8) and (15) one gets

$$\Gamma_0(\mathbf{p},\mathbf{p}';K)=\frac{\Phi_m^{(0)}(\mathbf{p})\Phi_m^{(0)*}(\mathbf{p}')}{\Omega-\Omega_{\mathbf{K}}^m+i\delta},$$

$$\Gamma(\mathbf{p},\mathbf{p}';K)=\frac{\Phi_m^{(0)}(\mathbf{p})\Phi_m^{(0)*}(\mathbf{p}')}{\Omega-\widetilde{\Omega}_{\mathbf{K}}^m}. \tag{20}$$

The substitution of the expressions (20) in the Bethe-Salpeter Eqs. (7) and (11) leads after some algebra to the final relationship

$$\widetilde{\Omega}_{\mathbf{K}}^m-\Omega_{\mathbf{K}}^m=\sum_{\mathbf{p}}\Phi_m^{(0)}(\mathbf{p})[\widetilde{G}(\mathbf{p},\mathbf{K},\Omega_{\mathbf{K}}^m)-\widetilde{G}_0(\mathbf{p},\mathbf{K},\Omega_{\mathbf{K}}^m)]\Phi_m^{(0)*}(\mathbf{p}), \tag{21}$$

where $\Phi_m^{(0)}(\mathbf{p})=\widetilde{G}_0^{-1}(\mathbf{p},\mathbf{K},\Omega_{\mathbf{K}}^m)\Psi_m^{(0)}(\mathbf{p})=(-\epsilon_0^m-p^2/M_x)\Psi_m^{(0)}(\mathbf{p})$. The potential W_{12} does not enter Eq. (21) explicitly. The m binding energy ϵ_0^m completely determines the deuteron ground-state wave function $\Psi_m^{(0)}(\mathbf{p})$. This is a signature of the contact potential.

The polariton dispersion in Eq. (14) does not allow us to split off the center-of-mass motion in the bipolariton solutions given by Eqs. (17)–(19) and Eq. (21), respectively. As a result, the in-plane momentum $\hbar\mathbf{K}$ of the molecule influences the relative motion of two constituent x's and causes the $\mathbf{K}$-dependence of the m radiative corrections Δ^m and Γ^m. Furthermore, the bipolariton model recovers the enhancement factor $a_m^2\gg a_x^2$ of the giant oscillator strength model.

According to Eqs. (21) and (14), three decay processes contribute to the m radiative width $\Gamma^m(\mathbf{K})/2=-\Im m\{\widetilde{\Omega}_{\mathbf{K}}^m-\Omega_{\mathbf{K}}^m\}$. The first one is the dissociation of the quasi-2D molecule into two outgoing QW polaritons. This process satisfies the energy and in-plane momentum conservation $\widetilde{\Omega}_{\mathbf{K}}^m-\omega^{QW}(-\mathbf{p}+\mathbf{K}/2)-\omega^{QW}(\mathbf{p}+\mathbf{K}/2)=0$. The conservation of energy is well defined because within our model $\Im m\{\omega^{QW}\}=0$. In the second process ''$m\to$QW polariton (bulk light)+bulk light (QW polariton),'' the bulk outgoing photon leaves a recoil QW polariton. This decay is also allowed by the energy – in-plane momentum conservation $\widetilde{\Omega}_{\mathbf{K}}^m-\omega^{QW(\text{bulk})}(-\mathbf{p}+\mathbf{K}/2)-\omega^{\text{bulk}(QW)}(\mathbf{p}+\mathbf{K}/2)=0$. However, the imaginary term $(i/2)\Gamma^x_{\pm\mathbf{p}+\mathbf{K}/2}$ due to the finite radiative lifetime of the bulk states relaxes the energy conservation. The third channel is attributed to $\widetilde{\Omega}_{\mathbf{K}}^m-\omega^{\text{bulk}}(-\mathbf{p}+\mathbf{K}/2)-\omega^{\text{bulk}}(\mathbf{p}+\mathbf{K}/2)=0$. For the 3D bipolariton model this process, which corresponds to the m decay into two outgoing polaritons of the upper dispersion branch, is strictly forbidden. However, the decay of the quasi-2D molecule into two bulk photons is weakly allowed due to the energy uncertainty given by $(1/2)(\Gamma^x_{\mathbf{p}+\mathbf{K}/2}+\Gamma^x_{-\mathbf{p}+\mathbf{K}/2})$.

Due to the pole structure of $\widetilde{G}(\mathbf{p},K)$ given by Eq. (14), one can easily discriminate Γ_{QW}^m (the first channel) and Γ_{bulk}^m (the second and third channels). From Eq. (21) we conclude

$$\Gamma_{QW}^m(\mathbf{K})+\Gamma_{\text{bulk}}^m(\mathbf{K})=(-1/2\pi^2)\Im m\{\int\Phi_m^{(0)}(\mathbf{p})\widetilde{G}(\mathbf{p},\mathbf{K},\Omega_{\mathbf{K}}^m)\Phi_m^{(0)}(\mathbf{p})d^2p\}.$$

In our experiments, the molecules with in-plane wave vector $K\simeq 0.55\times 10^5\ \text{cm}^{-1}\ll k_{\text{opt}}$ have been involved in transient m-mediated PL. The dependences of $\tau_{QW}^m=\hbar/\Gamma_{QW}^m(K\simeq 0)$ (solid line) and $\tau_{\text{bulk}}^m=\hbar/\Gamma_{\text{bulk}}^m(K\simeq 0)$ (dashed line) upon the exciton-photon coupling parameter R_c are plotted in Fig. 4. We conclude that indeed $\Gamma_{QW}^m\gg\Gamma_{\text{bulk}}^m$ within the 2D bipolariton model.

The dissociation of the molecule into two outgoing QW polaritons is more effective than the decay into the bulk modes due to (i) the x-weight factors $(\varphi^{QW}\varphi^{QW})>(\varphi^{QW}\varphi^{\text{bulk}})$ in the numerators of Eq. (14) and (ii) the joint density of states determined by the denominator of Eq. (14), i.e., by the functions $\delta\Omega^{\alpha,\alpha'}(\mathbf{p},\mathbf{K})=\widetilde{\Omega}_{\mathbf{K}}^m-\omega^{\alpha}(-\mathbf{p}+\mathbf{K}/2)-\omega^{\alpha'}(\mathbf{p}+\mathbf{K}/2)$. The joint density of QW polariton states given by $J^{QW,QW}=(1/4\pi^2)\int d^2p\,\delta[\widetilde{\Omega}_{\mathbf{K}}^m-\omega^{QW}(\mathbf{p}+\mathbf{K}/2)-\omega^{QW}(-\mathbf{p}+\mathbf{K}/2)]$ is considerably higher than the joint density of states for the m decay into the bulk modes. Moreover, the energy-momentum conservation surface $\delta\Omega^{QW,QW}(\mathbf{p},\mathbf{K})=0$ in 3D momentum space (p_x,p_y,K) shown in Fig. 5 has a critical saddle point O at $\mathbf{K}_0\simeq 2\mathbf{k}_{\text{opt}}$. The critical point is characterized by $\partial\delta\widetilde{\Omega}(\mathbf{p},\mathbf{K})/\partial p_x=\partial\delta\widetilde{\Omega}(\mathbf{p},\mathbf{K})/\partial p_y=0$ and gives rise to a logarithmic van

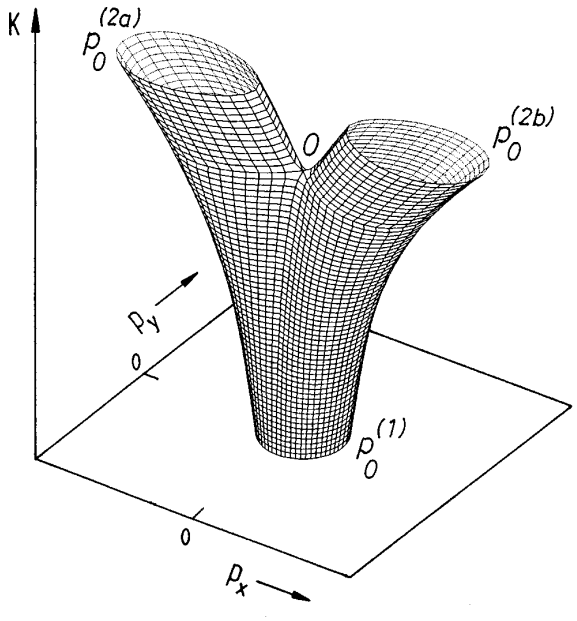

FIG. 5. The surface $\delta\Omega^{QW,QW}(\mathbf{p},\mathbf{K})=\tilde{\Omega}^m_{\mathbf{K}}-\omega^{QW}(\mathbf{p}+\mathbf{K}/2)-\omega^{QW}(-\mathbf{p}+\mathbf{K}/2)=0$ in 3D momentum space (p_x, p_y, K). The point O ($p_x=p_y=0$, $K=K_0$) is the critical saddle point. The wave vector $\mathbf{K}_0$ corresponds to the degenerate two-QW-polariton resonant decay of the molecule with the in-plane wave vector $\mathbf{K}_0$ into σ^+ and $+\sigma^-$ interface polaritons with the wave vector $\mathbf{K}_0/2$.

Hove singularity in the joint density of QW polariton states. This singularity, which is absent for the m decay into the bulk modes, is responsible for the strong coupling of the quasi-2D molecule with the interface (surface) light. One can trace the influence of the critical point at $\mathbf{K}_0$ up to the small in-plane m wave vectors $\mathbf{K}\simeq 0$.

III. DISCUSSION

The 2D bipolariton model shows that $\tau^m_{QW}\leqslant\tau^x_c$. Therefore, the excitonic molecules dissociate already within the initial transient stage, while the following exponential decay of the cross-polarized PL [see Figs. 1(a) and 1(b)] is due to escape of secondary QW polaritons into the bulk modes. The complete kinetics of the cross-polarized PL is approximated by

$$I_\perp(t)=I_0\{[e^{-t/\tau^m_{QW}}+1-2e^{-t/2\tau^m_{QW}}\cos(\epsilon^m_0 t)]+2\rho(1-e^{-t/\tau^m_{QW}})\}e^{-t/\tau^x}, \tag{22}$$

where $\tau^x\gg\tau^m_{QW}$ is the characteristic decay time of PL at the second exponential stage and ρ is a parameter which characterizes the fraction of the secondary QW polaritons escaped into the bulk modes. These secondary polaritons are due to the quick dissociation of the quasi-2D molecules.

A fit of the cross-polarized PL within the bipolariton kinetics of Eq. (22) is shown in Figs. 1(a) and 1(b) (solid line) for samples no. 1 ($d=130$ Å) and no. 3 ($d=200$ Å), respectively. We get the following parameters: $\tau^m_{QW}=0.5$ ps, $\epsilon^m_0=1.8$ meV, $\tau^x=27.4$ ps, $\rho=0.7$ for no. 1 and $\tau^m_{QW}=0.8$ ps, $\epsilon^m_0=0.7$ meV, $\tau^x=36.6$ ps, $\rho=0.3$ for no. 3. The m contribution to the complete time-resolved PL is also plotted in Figs. 1(a) and 1(b) (dashed line). Although the m optical decay into the bulk modes is relatively weak ($\tau^m_{\text{bulk}}\gg\tau^m_{QW}$), this channel yields the m-mediated PL signal in our experiments and tests the ''hidden'' dissociation of molecules into outgoing interface polaritons. In order to get a direct evidence of the hidden channel, one should visualize the dissociation of quasi-2D m's into QW polaritons by the QW grating technique[27] or by the detection of the light from the GaAs QW's edges (this technique has been developed in Ref. 32).

Using the dependence $\Gamma^m_{QW}=\Gamma^m_{QW}(R_c)$ of Fig. 4, we estimate $\hbar^2R_c\simeq 25\times10^{-2}$ eV^2 Å for sample no. 1 and $\hbar^2R_c\simeq 18\times10^{-2}$ eV^2 Å for no. 3. The increase of ϵ^m_0 and R_c with decrease of the thickness d from 200 Å (no. 3) to 130 Å (no. 1) follows the theoretical conclusions of Ref. 33 and Refs. 25 and 26, respectively. Moreover, the 2D bipolariton model with $\tau^m_{QW}<\tau^x_c$ explains the experimental results[10] on four-wave mixing in GaAs QW's ($d=100$ Å) where the m-mediated signal decays with $\tau^m\simeq 0.8$ ps, while the x dephasing time $\tau^x_c\simeq 3.9$ ps$\gg\tau^m$.

The width of the Lorentz spectral band of secondary QW polaritons is determined by the energy uncertainty $\Gamma^m(K\simeq 0)$ in the generation sequence two incoming photons→virtual molecule→two outgoing QW polaritons. The center of the band is located at $\tilde{\Omega}^m_{\mathbf{K}=\mathbf{0}}/2$. A fraction of the secondary polaritons which belong to the x-like part of the QW polariton dispersion is $\Delta N/N\propto\int_{\epsilon^m/\Gamma^m}^{\infty}S_{QW}[dx/(1+x^2)]=S_{QW}[\pi/2-\arctan(\epsilon^m/\Gamma^m)]$, where $S_{QW}=\text{const}$ is the 2D density of QW x states. The secondary quasi-2D excitons, i.e., the x-like QW polaritons, can populate the radiative modes and contribute to the cross-polarized PL at $t>\tau^m_{QW}$. Estimates show that $(\Delta N/N)_{\text{sample no. 1}}\simeq(\Delta N/N)_{\text{sample no. 3}}$ within 20% accuracy. The secondary ''cold'' excitons equilibrate and completely thermalize with the lattice only in sub-ns scale.[16,17] The measured decay of the thermalized x's in samples nos. 1–3 also agrees with the earlier measurements.[15]

The secondary quasi-2D excitons from the narrow frequency band $\omega_t\leqslant\omega\leqslant\omega_t+\Gamma^x$ couple resonantly with the radiative modes $p\leqslant k_{\text{opt}}$. The width of the band is due to the energy uncertainty $\Gamma^x(\Gamma^x\ll\Gamma^m\simeq\Gamma^m_{QW})$ of the radiative modes. The umklapp between the bulk and confined modes is due to Rayleigh scattering by QW impurities and interface fluctuations as well as due to Brillouin scattering by low-energy LA- and TA- phonons. The efficiency of the optical escape of secondary QW polaritons is proportional to the population of the bulk modes and inversely proportional to the x radiative lifetime within the photon cone. As a result, the parameter ρ of Eq. (22) is $\rho\propto\langle\Gamma^x_{\mathbf{p}}\rangle^2\propto R_c^2$. Here, the x radiative rate $\Gamma^x_{\mathbf{p}}$ is given by Eq. (4). The values of the coupling parameter R_c estimated from the transient stage at $t\leqslant\tau^m_{QW}$ yield $(R_{c,\text{sample no. 1}})^2/(R_{c,\text{sample no. 3}})^2\simeq 1.9$, while the fit of the total kinetics by Eq. (22) gives $\rho_{\text{sample no. 1}}/\rho_{\text{sample no. 3}}\simeq 2.3$. The coincidence of the numerical values is reasonable. Moreover, the characteristic decay time of the cross-polarized PL at $t>\tau^m_{QW}$ is $\tau^x\propto\langle\Gamma^x_{\mathbf{p}}\rangle^{-1}\propto R_c^{-1}$. Again, the ratio $\tau^x_{\text{sample no. 3}}/\tau^x_{\text{sample no. 1}}\simeq 1.3$ is quite well reproduced by the ratio $R_{c,\text{sample no. 1}}/R_{c,\text{sample no. 3}}\simeq 1.4$ found from the initial transient.

The resonant scattering of the incident light by interface disorder can also lead to an initial transient in exciton PL with the rise time given by the inverse of the inhomogeneous width.[34] The extremely short rise time of the m-mediated PL $[\tau(I_{\perp}^{\max}) \leqslant 1.5$ ps for the all studied samples], however, rules out the possibility that the observed rise time is due to interface disorder. According to our measurements, the typical width $(\delta\omega)_x$ of the x PL line is less than 0.2 meV corresponding to a time scale longer than 5 ps. Formally, the radiative corrections Δ^m and Γ^m to the quasi-2D m binding energy ϵ_0^m is considerably large than $(\delta\omega)_x$.

The dimensionless parameter $\delta=(\tau_{QW}^m \epsilon_0^m)^{-1} < 1$ determines an accuracy of the estimate of the m binding energy ϵ_0^m by Eq. (2). For $\delta \ll 1$, Eq. (2) gives a precise value of ϵ_0^m. In our case, $\delta \sim 0.4-0.5$ and Eq. (2) yields $\epsilon_0^m = 2.4$ meV for sample no. 1 and $\epsilon_0^m = 1.4$ meV for no. 3. These values are about 20–50 % large than the corresponding values obtained by the fitting procedure from Figs. 1(a) and 1(b).

The initial transient of the m-mediated PL after an impact optical excitation should also occur in bulk semiconductors. For example, our 100 fs time-resolved technique is well suited for bulk CdS or CdSe, where $\epsilon_0^m \simeq 3-5$ meV. Because the 3D bipolariton model gives $\delta \ll 1$, one anticipates to see a few complete beats in the transient m-mediated PL. Moreover, in this case the estimate of ϵ_0^m by Eq. (2) should be very precise.

The extremely fast dissociation of the molecules into QW polaritons with $\tau_{QW}^m \leqslant \tau_c^x$ shows that the complete dynamical sequence of the 2D bipolariton model, i.e., two incoming photons σ^+ and $\sigma^- \rightarrow$two virtual excitons σ^+ and $\sigma^- \rightarrow$molecule (bipolariton)$\rightarrow$two outgoing QW polaritons σ^+ and σ^-, holds as a conservative, coherent process. Such a behavior, which shows that the coupling of the molecule with the light dominates over the scattering processes, is the fundamental signature of the bipolariton model. For example, this property occurs for biexcitons in bulk CuCl at $T \leqslant 10$ K due to an anomalously weak $m(x)$ LA-phonon coupling.[35] In our experiments with high-quality GaAs QW's, this feature also holds because the m-mediated transient PL is independent of the temperature $T \leqslant 30$ K and the intensity of the optical pump. In other words, an optically generated molecule exists in a ''frozen'' state with the conserved in-plane momentum $\hbar\mathbf{K}$ and radiatively decays before scattering or dephasing.

Within this picture, the m-mediated PL from QW's has a spatial anisotropy about in-plane projection $\mathbf{k}_{\|}$ of the wave vector of the pump light. Due to the unrelaxed in-plane momentum $\mathbf{K}=2\mathbf{k}_{\|}$ of the created quasi-2D molecules, the emission occurs predominantly in $\mathbf{k}_{\|}$ direction. This conclusion, which is valid for the m radiative decay into the both confined and bulk modes, stems directly from the conservation of the total in-plane momentum. A similar effect has been discussed for transient PL from a photogenerated electron-hole plasma in GaAs QW's.[36]

IV. CONCLUSIONS

In this work we analyze the transient coherent stage at the very beginning of the time-resolved m-mediated PL from GaAs QW's. The following conclusions summarize our study.

(i) The initial transient stage of the m-mediated photoluminescence is attributed to the coherent quantum evolution of σ^+ and σ^- excitons towards the m ground state right after the impact optical excitation $(\tau_{\text{pulse}} \ll \hbar/\epsilon_0^m)$. The finite rise time of PL provides us with a straightforward estimate of the molecule binding energy ϵ_0^m. The initial transient also reflects a type of quantum beats caused by the $x(\sigma^+)-x(\sigma^-)$ Coulombic attraction. These inherently decaying beats occur between the m ground state and the x-x dispersive continuum and cannot be analyzed within the standard few-level optical Bloch equations which simplify the description of the two-exciton continuum.

(ii) The 2D bipolariton model clarifies that the main ''hidden'' channel of the m radiative decay is the dissociation into two QW polaritons rather than the observable decay into the bulk modes with emission of photons outside a QW. We conclude that excitonic molecules dissociate already within the initial transient, while the following exponential decay of the cross-polarized PL is due to escape of secondary interface polaritons into the bulk modes. Moreover, due to the very short dissociation times $\tau_{QW}^m \sim 1$ ps, the optical generation of a quasi-2D molecule and the following radiative decay, i.e., the sequence two photons$\rightarrow$ two virtual excitons$\rightarrow$molecule$\rightarrow$two outgoing QW polaritons, is a coherent process.

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