Pis'ma v ZhETF, vol.65, iss.2, pp.164 - 169

INFLATING ANTENNA: DYNAMICS OF EXCITON WAVE PACKETS

E.I.Rashba

L. D. Landau Institute for Theoretical Physics
117940 Moscow, Russia

Department of Physics, University of Utah
UT 84112, USA

Submitted 15 December 1996

A theory of the free induction signal from biexcitons and bound excitons is presented. Simultaneous existence of the exciton continuum and a bound state is shown to result in a new type of the time dependence of the free induction. Optically detected signal increases in time and oscillates with increasing amplitude until damped by radiative and dephasing processes. Expanding area of a coherent exciton polarization (inflating antenna), produced by the exciting pulse, is the underlying physical mechanism. Developed formalism can be applied to different biexciton transients.

PACS: 42.50.-p, 71.35.-y

Ultrafast spectroscopy of excitons in the time domain [1] has proven to be a powerful tool to probe quantum coherence of exciton states which was originally studied by polarization of the stationary emission [2]. Most of the experimental data were taken from GaAs quantum wells, however, some experiments were performed with bulk excitons. Quantum beats in different response functions provide a manifestation of the coherence driven by external fields. These beats appear when several states having close energies are excited simultaneously. Quantum beats were observed with magnetically split exciton levels [3], bound excitons with different confinement energies [4], heavy and light hole excitons [5], free and bound excitons [6], and with biexcitons and a two-exciton continuum [7]. Two last examples are of principal importance for us in what follows. Experimental data provide convincing evidence of a strong effect of the exciton-exciton and exciton-free carrier interaction on nonlinear response functions [8, 9]. Different theoretical approaches were applied depending on the range of the parameter values. When nonradiative relaxation times τ are short, mean-field approach [10] works rather well. This paper deals with the opposite limit of the large τ region and is related to the papers Ref. [11, 12, 13] based on microscopic models.

The traditional approach to quantum beats is based on the energy spectrum comprising few, usually two, discrete energy levels. This approach can be applied to beats between heavy and light hole excitons because of the momentum conservation and absence of the interaction between these excitons. However, the biexciton and bound exciton problems are more involved because of the existence of the two-exciton and single-exciton continuum, respectively. E.g., for a two-exciton system it is the exciton-exciton interaction which supports the two-photon coherence, and the lower part of the continuum with the width about several biexciton binding energies, ε_b, contributes to the coherent polarization along with the bound biexciton state. In addition to the theoretical arguments, some experimental data provide weighty, although indirect, evidence of the role of the two-exciton continuum.

Indeed, it was shown [11] that the four-level biexciton energy scheme can be brought into agreement with experimental data only if the enhancement factor typical of giant oscillator strengths [14] is invoked. More recent data [15] provide evidence of the significant role of the exciton-exciton interaction in the continuum. Therefore, frequencies of allowed transitions are distributed continuously and the spread of the frequencies is about ε_b. Naive consideration suggests that such an energy spectrum should result in beats having the frequency of about ε_b and showing fast nondissipative decay because of accumulating phase differences between different modes. It turns out that the actual physical picture is quite different.

We present an exact solution for a free induction signal excited by a single-side exponential pulse, $t < 0$, in a nondissipative system with a biexciton nonlinearity. The special shape of the pulse simplifies calculations but does not influence the basic results. The contribution of the two-exciton continuum is consistently taken into account. With such an approach, free induction, i.e., free oscillations of a two-exciton wave function $\Psi(t)$ for $t > 0$, includes two modes. There exists *a beating mode* describing *undamped beats whose frequency is equal to* ε_b. There exists also *a growing mode* whose amplitude increases linearly with time, t, and whose carrier frequency equals the energy of the bottom of the two-exciton continuum. The growing mode is inherent in interacting systems possessing a continuous spectrum. It describes the inflation in a real space of the wave packet created by the pulse. The two modes result in *an optically detected free induction signal which increases with t and has a monotonic and oscillating parts.* Increase of the signal is restricted by the strong *radiative decay* resulting in short emission pulses. The same modes exist for excitons bound to impurities. We expect that these modes contribute also to different nonlinear processes, including multiple-impulse processes, and that the developed technique is of general applicability.

To make clear the basic idea and account rigorously for analytical properties of the exciton Green functions, we develop an exactly soluble model. To this end we neglect polariton effects and dephasing. We also neglect the dependence of the scattering amplitude on the light polarization because it is sensitive to the band structure, geometry, etc. [9, 16]. Excitons are considered as stable particles without internal degrees of freedom. It is convenient to start with the bound exciton problem. If the electromagnetic wave $\mathbf{E}_\mathbf{q}(\mathbf{r},t) \propto \exp\{i(\mathbf{q}\cdot\mathbf{r} - \omega t) + \alpha t\}$, $\alpha > 0$, is incident upon a crystal at $-\infty < t < 0$, the exciton wave function $\Psi_\mathbf{q}(\mathbf{q}',t)$ at the instant t, $t > 0$, can be calculated as a linear response to this perturbation

$$\Psi_\mathbf{q}(\mathbf{q}',t) = (iM/\sqrt{v})A_{\mathbf{q}'\mathbf{q}}(t) \ , \quad A_{\mathbf{q}'\mathbf{q}}(t) = \sum_j \frac{\psi_j(\mathbf{q}')\bar{\psi}_j(\mathbf{q})}{\omega - E_j + i\alpha} e^{-iE_j t} \ . \qquad (1)$$

Here $\Psi_\mathbf{q}(\mathbf{q}',t)$ and $\psi_j(\mathbf{q})$ are, respectively, time-dependent and stationary exciton wave functions in the momentum representation. The subscript $j \geq 1$ numerates single-exciton states, bound and free ones. Ground state energy of the crystal is chosen as the origin, $E_0 = 0$. The coefficient M is the matrix element, per unit cell, of the perturbation produced by the field $\mathbf{E}(\mathbf{r},t)$, and v is the unit cell volume. The momenta $\mathbf{q}$ and $\mathbf{q}'$, which are of importance for optical experiments, are small and will be neglected in the final results.

Amplitude $A_{\mathbf{q}'\mathbf{q}}(t)$ describes free precession of $\Psi_{\mathbf{q}}(\mathbf{q}', t)$ for positive times. For $t = 0$, $A_{\mathbf{q}'\mathbf{q}}(t)$ coincides with the retarded exciton Green function

$$G_{\mathbf{q}'\mathbf{q}}(\omega) = \sum_j \psi_j(\mathbf{q}')\bar{\psi}_j(\mathbf{q})/(\omega - E_j + i0) \tag{2}$$

for $\omega \to \omega + i\alpha$. For arbitraty t, functions $A_{\mathbf{q}'\mathbf{q}}(t)$ and $G_{\mathbf{q}'\mathbf{q}}(\omega)$ are related by the equation

$$A_{\mathbf{q}'\mathbf{q}}(t) = \frac{1}{2\pi i}\int_{-\infty}^{\infty} d\omega' \ \frac{\exp(-i\omega' t)}{\omega' - \omega - i\alpha} G_{\mathbf{q}'\mathbf{q}}(\omega') . \tag{3}$$

Eq. (3) can be checked by emloying (2), closing the integration path in the lower complex half-plane, and calculating residues in the poles of $G_{\mathbf{q}'\mathbf{q}}(\omega')$.

Subsequent transformations of $A_{\mathbf{q}'\mathbf{q}}(t)$ are based on the introduction of the scattering operator $\hat{T}$ [17]

$$G_{\mathbf{q}'\mathbf{q}}(\omega) = G^0_{\mathbf{q}}(\omega)\delta_{\mathbf{q}'\mathbf{q}} + G^0_{\mathbf{q}'}(\omega)T_{\mathbf{q}'\mathbf{q}}(\omega)G^0_{\mathbf{q}}(\omega) , \tag{4}$$

where $G^0_{\mathbf{q}}(\omega) = (\omega - \varepsilon(\mathbf{q}) + i0)^{-1}$ is a free-exciton Green function. Only the second term of Eq. (4) contributes to $A_{\mathbf{q}'\mathbf{q}}(t)$ for $\mathbf{q}' \neq \mathbf{q}$ and will be retained below. It is an important property of this term that it includes a product of two G^0 functions with nearly coinciding poles. This property strongly influences the subsequent results. $T_{\mathbf{q}\mathbf{q}'}(\omega)$ is analytical in the upper half-plane, hence, the following dispersion relation holds for it

$$\hat{T}(\omega) = -\frac{1}{\pi}\int_{-\infty}^{\infty} d\omega' \ \hat{T}''(\omega')/(\omega - \omega' + i0) , \tag{5}$$

where $\hat{T}''(\omega) = \mathrm{Im}\{\hat{T}(\omega)\}$. Sustituting (4) and (5) into (3) and performing integration over ω', one gets in the $\mathbf{q}', \mathbf{q} \to 0$ limit:

$$A(t) = -\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \ \frac{d}{d\varepsilon}\left[\frac{T''(\omega')}{\omega' - \varepsilon}\left(\frac{\exp(-i\omega' t)}{\omega - \omega' + i\alpha} - \frac{\exp(-i\varepsilon t)}{\omega - \varepsilon + i\alpha}\right)\right] . \tag{6}$$

Here $\varepsilon = \varepsilon(0)$ is the energy of long wave-length excitons, and $A(t)$ and $T''(\omega')$ are the limits of $A_{\mathbf{q}'\mathbf{q}}(t)$ and $T_{\mathbf{q}'\mathbf{q}}(\omega)$, respectively, for $\mathbf{q}, \mathbf{q}' \to 0$. Eq. (6) is the final equation for the time dependent amplitude $A(t)$. It is completely determined by the operator $\hat{T}$.

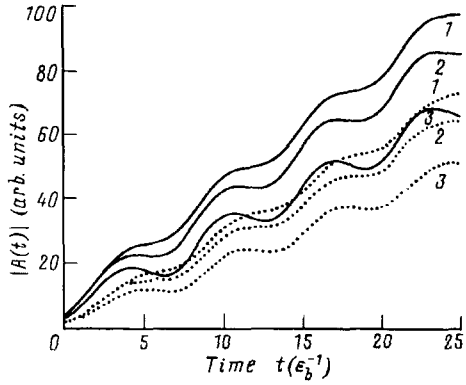
Two basic properties of Eq. (6) follow from general arguments.

First, the derivative $d/d\varepsilon$ results in a contribution to $A(t)$ proportional to $t\exp(-i\varepsilon t)$. The term in $A(t)$ increasing with t will be termed below as *the growing mode*. It originates from the product of two G^0 functions with coinciding poles, Eq. (4). The growing mode is reminiscent of the growing solutions of differential equations with degenerate characteristic numbers. This mode describes the global evolution of the wave packet prepared by the pulse. As t increases, the packet expands in r space. For translationally invariant systems this expansion is accompanied by changes in phases of different Fourier components, whereas their moduli remain unchanged. However, the impurity potential violates the momentum conservation, and the amplitude of the $\mathbf{q} = 0$ mode increases with t. The giant oscillator strengths observed in stationary experiments are ascribed to *exciton*

antennas [18]. In these terms *the growing mode is an inflating exciton antenna.* This picture explains why the growing mode is specific for systems possessing a continuous spectrum and, hence, extended states.

Second, the bound state is a pole of $\hat{T}(\omega)$. Therefore, $T''(\omega')$ includes a term proportional to $\delta(\omega' - \varepsilon + \varepsilon_b)$. It contributes into Eq. (6) an oscillating exponent $\exp[-i(\varepsilon - \varepsilon_b)t]$. The exciton band width is supposed to be large as compared with ε_b. Under these conditions the integration along the cut in the complex plane contributes the factor $\exp(-i\varepsilon t)$. Two oscillating terms in Eq. (6), $\exp[-i(\varepsilon - \varepsilon_b)t]$ and $\exp(-i\varepsilon t)$, result in beats at the frequency of ε_b with a time independent amplitude. This oscillating contribution to $A(\omega)$ will be termed as *the beating mode.*

The integral of Eq. (6) can be easily performed for a Frenkel exciton when the impurity potential is described by a degenerate perturbation $U_{\mathbf{mn}} = -U\delta_{\mathbf{m}0}\delta_{0\mathbf{n}}$[19]. Here m and n numerate lattice sites, and the impurity resides at the site $\mathbf{m} = \mathbf{n} = 0$. In this case $T_{\mathbf{qq}'}(\omega)$ does not depend on the momenta q and q'. For a two-dimensional (2D) system, the density-of-states inside the exciton band can be chosen as $\rho(\omega) = 1/E_B$, where E_B is the band width. The amplitude $A(t)$ is shown in Fig. for three values of ω. Both the linear-in-t growth and the oscillations with a time independent amplitude are distinctly seen in the asymptotic region, $t2\pi\varepsilon_b^{-1}$. Actually, they are seen even for small values of $t > 0$, but the shape of the first oscillation is somewhat distorted. It also depends on the shape of the exciting pulse. The data for a 3D system with $\rho(\omega) = 8\sqrt{\omega(E_B - \omega)}/\pi E_B{}^2$ are also shown in Fig. 1. The dependence of $A(t)$ on dimensionality is rather weak.



Time dependence of the amplitude $|A(t)|$ for $\alpha = \varepsilon_b$, $E_B = 10\varepsilon_b$. 3D – solid lines, 2D – dotted lines. (1) $\omega - \varepsilon = 0$, (2) $\omega - \varepsilon = -0.5\varepsilon_b$, (3) $\omega - \varepsilon = -\varepsilon_b$. For $t2\pi/\varepsilon_b$ the amplitude shows a linear growth and undamped oscillations with a period $2\pi/\varepsilon_b$

Therefore, after a short transient the growing and beating modes dominate the amplitude $A(t)$. The optically detected free-induction signal, $I(t)$, is related to the zero-momentum component of the wave function. Therefore, $I(t) \propto |\Psi_0(0,t)|^2$. In the asymptotic region $A(t) \propto \{t + \frac{b}{2}\exp[i(\varepsilon_b t + \phi)]\}e^{-i\varepsilon t}$, where b and ϕ are real parameters, and $I(t)$ obeys the law:

$$I(t) \propto \{t^2 + bt\cos(\varepsilon_b t + \phi)\}. \tag{7}$$

Interference of the two modes results in an unusual shape of the signal $I(t)$. It consists of the monotonic and oscillatory contributions, and both of them growing (rather than decaying!) with t. If to neglect the oscillating part in (7), the

radiative time $\tau_R(t)$ decreases with t as $\tau_R(t) \propto t^{-2}$. This rapid increase in the emission probability establishes the applicability limit for Eq. (7). One can infer from the data of Fig. 1 that $\tau_R^{-1}(t) \approx (\tau_R^0)^{-1}(1+\beta\varepsilon_b t)^2$, where $\beta \sim 1$, and τ_R^0 can be estimated as the bound-exciton radiative lifetime. Radiative lifetime $\tau_{\rm em}$ can be evaluated from the phenomenological equation

$$\int_0^1 dn = \int_0^{\tau_{\rm em}} dt/\tau_R = 1 \ , \tag{8}$$

which results in $\tau_{\rm em} \approx (\tau_R^0/\varepsilon_b^2)^{1/3}$. This estimate is crude because $A(t=0)$, Eq. (1), depends on ω and α. For $\tau_R^0 \approx 1$ ns and $\varepsilon_b \approx 10$ meV, we get $\tau_{\rm em}\varepsilon_b \approx 25$, which correspons to about three oscillations in $|A(t)|$, Fig. 1. Therefore, the radiative response has a shape of a short train of oscillations with the total duration of only about $\tau_{\rm em}$ and the efficiency up to 100%. Polariton effects neglected above are expected to contribute at this fast stage of the radiative decay, and this contribution should be dimensionality dependent [20].

In what follows we generalize these results for biexcitons. There exist two processes which result in optical production of biexcitons [7, 14]. The first process is two-step absorption with an exciton level as a real intermediate state. In this process an exciton produced at the first step act as an "impurity". All above results are applicable to this process without any serious changes. The second process is two-phonon absorption from the ground state. The theory of this process is more cumbersome than for impurity absorption. Nevertheless, the final results are nearly identical.

Biexciton eigenfunctions can be written in the operator form as

$$|{\rm K}j\rangle = \frac{1}{\sqrt{2}} \int \frac{d{\rm k}}{(2\pi)^3} \ \psi_j({\rm k}) \ \psi^\dagger_{{\rm K}/2+{\rm k}} \psi^\dagger_{{\rm K}/2-{\rm k}} \, , \tag{9}$$

where $\psi^\dagger_{{\rm K}/2\pm{\rm k}}$ are exciton creation operators, and K is the center-of-mass momentum of a biexciton. Functions $\psi_j({\rm k})$ are eigenfunctions in the momentum representation. The biexciton wave function at positive times can be found in the second order of the perturbation theory in the field $\mathbf{E}_{\mathbf{q}}(\mathbf{r},t)$. In the momentum representation

$$\Psi_{\mathbf{q}}({\rm k},t) = \frac{2M^2\sqrt{V}/v}{\omega - \varepsilon({\rm q}) + i\alpha} A_{k0}(t) \ , \quad A_{k0}(t) = \sum_j \frac{\psi_j({\rm k})\bar{\psi}_j(0)}{2\omega - E_j + 2i\alpha} \ {\rm e}^{-iE_j t} \, . \tag{10}$$

Here E_j are the energy levels of a two-exciton system, and V is the normalization volume; momentum ${\rm K} = 2{\rm q}$. If biexcitons are excited by two light beams with momenta $\mathbf{q}_1$ and $\mathbf{q}_2$, function $\bar{\psi}_j(0)$ in (10) should be substituted by $\bar{\psi}_j((\mathbf{q}_1 - \mathbf{q}_2)/2)$.

Eq. (10) differs from Eq. (1) only in the coefficient and in change in variables, $\omega \to \Omega = 2\omega$ and $\alpha \to 2\alpha$. Therefore, the transformations which led us from Eq. (1) to Eq. (6) can be repeated for biexcitons step by step. The equation for the scattering operator $T_{\mathbf{k}'\mathbf{k}}(\Omega)$ depends on the interaction between excitons. The zero-radius potential provides a satisfactory approximation for giant oscillator strengths [14]. With this potential, the operator $\hat{T}(\Omega)$ is known for three-dimensional systems [17], and it does not depend on momenta. Finally, $A(t)$ shows actually the same behavior as for bound excitons.

One can infer from Eq. (9) that the quantum state Ψ_q decays into two photons with momenta $K/2 \pm k$. Intensity of the free induction signal is proportional to $|\Psi_q(k,t)|^2 \approx |\Psi_0(0,t)|^2$. In the asymptotic region $A(t) \propto \{t + \frac{b}{2}\exp[i(\varepsilon_b t + \phi)]\}\exp(-2i\varepsilon t)$, and Eq. (7) describes the optically detected signal.

In conclusion, coexistence of the continuous spectrum and a bound state results in existence of growing and beating modes in the free induction $\Psi(t)$, following the exciting pulse, for biexcitons and bound excitons. The duration of the induction signal is controlled by the radiative decay rate and dephasing. If the first mechanism dominates, the signal is emitted in a short pulse with the radiative yield close to unity.

I am grateful to M. D. Sturge and J. M. Worlock for suggestive discussions and critical reading the manuscript. The support of the Office of Naval Research under Contract No. N000149410853 is acknowledged.

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