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Weak localization of exciton polaritons in a quantum well

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We describe effects of weak localization of exciton polaritons (EP's) and induced scattering of EP's into the zero-wave-vector mode in a planar semiconductor microcavity containing quantum wells (QW's). Coherent light emission in the direction normal to the surface under the tilted pumping can be explained in terms of the weak localization of EP's in QW's. [S0163-1829(96)51228-2]

The enhancement of backscattering on the linear propagation of waves in a dense distribution of elastic scatterers has been recognized almost independently in two different fields. In condensed-matter physics, this backscattering leads to the weak-localization regime for electrons in impure metals, which led to the dimensional dependence of Anderson localization.[1,2] In optics, the pioneering work of de Wolf and others showed that light scattered from disordered dielectrics can show a polarization-dependent sharp peak in the intensity in the backscattered direction.[3–9] The effect of weak localization of waves on the nonlinear optical properties of dielectric media was first investigated by one of the authors[10] in the context of the enhancement of phase conjugation due to coherent backscattering of excitons or exciton polaritons (EP's). A number of other theoretical discussions of weak localization effects in nonlinear optics have been published,[11–14] but no experimental observation of these effects has yet been reported.

Recently, however, Rhee *et al.* observed the transfer of coherence between EP states in a system consisting of multiple-quantum-well excitons resonantly coupled to a planar Fabry-Pérot microcavity.[15] Specifically, they observed the emission of light in the direction perpendicular to the surface with a small divergence angle, even though the EP states were excited at an angle $(3°)$ away from the normal. The emitted light was coherent with the incident pump light (determined by interfering the emission with the pump beam), and the intensity increased approximately as the square of the pump intensity. These experimental observations can be accounted for as a consequence of exciton-exciton interactions in a weakly localized system in the following way. The pump excites EP with an initial in-plane wave vector $\mathbf{k}_0$. These EP's may be coherently backscattered due to disorder in the quantum-well confinement potential into states with momentum $-\mathbf{k}_0$. Collisions between EP with momentum $+\mathbf{k}_0$ and $-\mathbf{k}_0$ result in generation of a population of EP at exactly $\mathbf{k}=\mathbf{0}$, giving rise to coherent emission of light in the normal direction. In this paper we develop a theoretical model for interacting EP's in a disordered system, showing how the observations of Rhee *et al.* may be understood as a consequence of weak localization of EP. Further consequences of the weak localization are also presented which should be amenable to experimental test.

We begin by assuming that the incident laser tilted at an angle θ from the normal excites a coherent state $|\alpha\rangle$ of the EP with an in-plane momentum $\mathbf{k}_0$ in a quantum well. The annihilation and creation operators for the initially excited EP with momentum $\mathbf{k}_0$ are c_α and $c_\alpha^\dagger$, and those for zero momentum (the signal mode) are c_s and $c_s^\dagger$. The emission intensity in the normal direction may be considered to be proportional to the number of EP's generated in the signal mode per second, and may be evaluated under cw excitation as

$$I_s=\lim_{t\to\infty}\frac{\partial}{\partial t}\langle\langle c_s^\dagger c_s\rho(t)\rangle\rangle. \tag{1}$$

The double angular brackets in (1) signify both the quantum-mechanical average and ensemble average over the distribution of scattering potentials, which comes mainly from the fluctuation in quantum-well width in the present case. An irrelevant factor coming from transmissivity (coupling of the polaritons to the external electric field) has been omitted. The density operator $\rho(t)$ of the total system is described as follows:

$$\rho(t)=\exp(-iH_Tt)\rho_0\exp(iH_Tt), \tag{2}$$

where

$$H_T=H+H'+H'', \tag{3}$$

$$H=\sum_{\mathbf{k}}\omega(\mathbf{k})c_{\mathbf{k}}^\dagger c_{\mathbf{k}}+\sum_{\mathbf{kq},i}V_0(\mathbf{q})e^{i\mathbf{q}\cdot\mathbf{r}_i}c_{\mathbf{k}+\mathbf{q}}^\dagger c_{\mathbf{k}}, \tag{3a}$$

$$H'=\sum_\alpha(c_\alpha+c_\alpha^\dagger)\boldsymbol{\mu}_\alpha\cdot\mathbf{E}, \tag{3b}$$

$$H''=\sum_{\mathbf{k}}V_{\mathbf{k}}c_s^\dagger c_d^\dagger c_\alpha c_{\mathbf{k}}+\text{H.c.} \tag{3c}$$

Here and hereafter we put $\hbar=1$. The radiation field has been divided into two parts: inside and outside the microcavity. The former is taken into account in the EP states $(c_{\mathbf{k}},c_{\mathbf{k}}^\dagger)$, i.e., the hybrid exciton-field states in the cavity. The external field $\mathbf{E}$ excites the EP with wave vector $\mathbf{k}_0$ coherently, i.e., $c_\alpha|\alpha\rangle=\alpha|\alpha\rangle$. Here $\mathbf{k}_0$ is the two-dimensional wave vector of the EP in the quantum-well plane. This EP suffers from scattering by the random potential $V_0(\mathbf{r}_i)$, which comes mainly from the fluctuation of the well thickness around the point $\mathbf{r}_i$ in the quantum-well plane, and $V_0(\mathbf{q})$ is the Fourier component of $V_0(\mathbf{r})$ with wave vector $\mathbf{q}$. This random potential is written in terms of the well thickness fluctuation in space and the exciton-to-polariton transformation coefficients.[10] The coupling strength of external field $\mathbf{E}$ to the polaritons $(c_\alpha, c_\alpha^\dagger)$ in H' is determined by the transition dipole moment and the exciton-to-polariton transformation coefficients. The interaction given in H'' corresponds to scattering of an EP in the initial state $|\alpha\rangle$ with an EP of wave vector $\mathbf{k}$ to yield a ''signal'' EP (c_s) and an ''idler'' EP (c_d).

We will show, based on this Hamiltonian, that the initially excited EP's are scattered into the backward direction by the disorder potential, and the backward-propagating EP and the pumped EP collide efficiently and coherently to produce a $\mathbf{k}=\mathbf{0}$ polariton which efficiently emits into the normal direction, as discussed in the Introduction. The formalism also allows the angular and temperature dependences to be predicted.

The scattering of EP's by the potential fluctuations $V_0(\mathbf{q})\exp(i\mathbf{q}\cdot\mathbf{r}_i)$ must be taken into account to infinite order to evaluate the effects of the weak localization of EP's. Therefore the Hamiltonian H of (3a) is kept to the final stage of the calculation, where the ensemble average of observed physical variables is taken. The present signal appears only for the second and higher orders in H', i.e., the interaction with the external field, and also in H'', i.e., the EP-EP collision. Therefore we expand the density operator $\rho(t)$ of (1) in H' and H'', and we keep the lowest order terms with finite contribution to the signal:

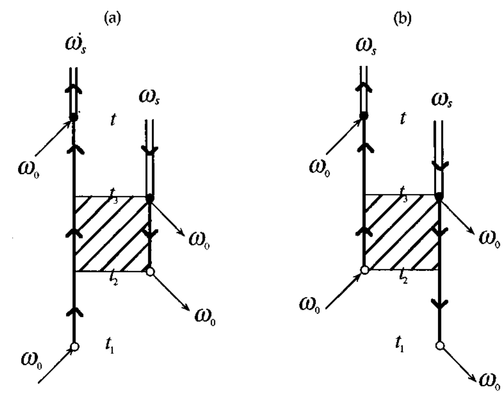

FIG. 1. Two diagrams which contribute to the emission into the direction normal to the surface under the laser pumping tilted from normal incidence. Vertical lines describe propagation of the exciton polariton, the signal (ω_s) and the idler (ω_d) polaritons and slanted lines the coherent incident polariton $(\omega_0, \mathbf{k}_0)$.

$$\lim_{t\to\infty}\frac{\partial}{\partial t}\langle\langle c_s^\dagger c_s\rho(t)\rangle\rangle=\lim_{t\to\infty}\int_{-\infty}^{t}dt_3\int_{-\infty}^{t_3}dt_2\int_{-\infty}^{t_2}dt_1(-i)^4\langle\langle c_s^\dagger c_s[H''(t),[H''(t_3),[H'(t_2),[H'(t_1),\rho_0]]]]\rangle\rangle. \tag{4}$$

This finite contribution comes from two kinds of processes shown in Fig. 1. Then the integrand is rewritten in the following terms:

$$A\{\langle G_1(t,t_3,t_2,t_1)\rangle+\langle G_2(t,t_3,t_2,t_1)\rangle\}, \tag{5}$$

with

$$A=\langle 0,\alpha|c_d c_s H''|\mathbf{k},\alpha\rangle\langle\alpha|c_\alpha^\dagger\boldsymbol{\mu}\cdot\mathbf{E}|\alpha\rangle\langle\alpha|c_\alpha\boldsymbol{\mu}\cdot\mathbf{E}|\alpha\rangle \times\langle\mathbf{k},\alpha|H''c_s^\dagger c_d^\dagger|0,\alpha\rangle, \tag{6}$$

$$G_1=(-i)^4 e^{i(\omega_s+\omega_d-\omega_0)(t-t_3)}e^{i\omega_0(t_2-t_1)} \times\langle\mathbf{k}|e^{-iH(t-t_1)}|\mathbf{k}_0\rangle_+\langle\mathbf{k}_0|e^{iH(t_3-t_2)}|\mathbf{k}\rangle_-, \tag{7a}$$

$$G_2=(-i)^4 e^{i(\omega_s+\omega_d-\omega_0)(t-t_3)}e^{-i\omega_0(t_2-t_1)} \times\langle\mathbf{k}|e^{-iH(t-t_2)}|\mathbf{k}_0\rangle_+\langle\mathbf{k}_0|e^{iH(t_3-t_1)}|\mathbf{k}\rangle_-. \tag{7b}$$

These Green's functions describe how the incident EP $\mathbf{k}_0$ is scattered into that with $\mathbf{k}$ nearly equal to $-\mathbf{k}_0$ by interference of multiple scattering with its time-reversed process, i.e., the weak localization. Here the coherently pumped EP with energy $\hbar\omega_0$ and wave vector $\mathbf{k}_0$ is obtained from the external field and constitutes the coherent state $|\alpha\rangle$. In (6), $|0\rangle$ denotes the vacuum state besides the pumped EP. The ensemble average of these Green's functions is approximated into the following form:

$$\langle G_1+G_2\rangle=(-i)^4 e^{i(\omega_s+\omega_d-\omega_0)(t-t_3)}\langle\langle\mathbf{k}|e^{-iH(t-t_3)}|\mathbf{k}\rangle_+\rangle \times\langle\langle\mathbf{k}|e^{-iH(t_3-t_2)}|\mathbf{k}_0\rangle_+\langle\mathbf{k}_0|e^{iH(t_3-t_2)}|\mathbf{k}\rangle_-\rangle \times\{e^{i\omega_0(t_2-t_1)}\langle\langle\mathbf{k}_0|e^{-iH(t_2-t_1)}|\mathbf{k}_0\rangle_+\rangle + e^{-i\omega_0(t_2-t_1)}\langle\langle\mathbf{k}_0|e^{iH(t_2-t_1)}|\mathbf{k}_0\rangle_-\rangle\}. \tag{8}$$

Here, assuming that the elastic scattering rate due to the potential fluctuation is much larger than that of the inelastic decay, the ensemble average of G_1 and G_2 given by (7a) and (7b) is approximated by the products of the ensemble averages of three propagators for time intervals $[t,t_3]$, $[t_3,t_2]$, and $[t_2,t_1]$. The subscript $\pm$ of the propagators means the upward and downward propagation of states in Fig. 1, and the outermost bracket means taking an ensemble average over distribution of scatterers. For time intervals $\tau=t-t_3$ and $\tau_1=t_2-t_1$, we have the retarded and advanced Green's functions:

$$G_{\mathbf{k}}^R(\tau)=-ie^{i(\omega_s+\omega_d-\omega_0)\tau}\langle\langle\mathbf{k}|e^{-iH\tau}|\mathbf{k}\rangle_+\rangle, \tag{9a}$$

$$G^R_{\mathbf{k}_0}(\tau_1) = -ie^{i\omega_0\tau_1}\langle\langle\mathbf{k}_0|e^{-iH\tau_1}|\mathbf{k}_0\rangle_+\rangle, \tag{9b}$$

$$G^A_{\mathbf{k}_0}(\tau_1) = -ie^{-i\omega_0\tau_1}\langle\langle\mathbf{k}_0|e^{iH\tau_1}|\mathbf{k}_0\rangle_-\rangle. \tag{9c}$$

The double Green's function is required for the time interval $\tau_2 = t_3 - t_2$:

$$\Xi(\tau_2) \equiv (-i)^2\langle\langle\mathbf{k}|e^{-iH\tau_2}|\mathbf{k}_0\rangle_+\langle\mathbf{k}_0|e^{iH\tau_2}|\mathbf{k}\rangle_-\rangle. \tag{10}$$

This can be well approximated by the sum of both ladder-type and maximally crossed diagrams, the latter of which brings about the weak localization. When we insert (9) and (10) into (8) and rewrite the integration in t_1, t_2, and t_3 in terms of τ, τ_1, and τ_2 in (4), we have the following expression:

$$I_s = AG^R(\omega_s+\omega_d-\omega_0,\mathbf{k})\Xi(\mathbf{k},\mathbf{k}_0) \times\{G^R(\omega_0,\mathbf{k}_0) - G^A(\omega_0,\mathbf{k}_0) + \text{c.c.}\}. \tag{11}$$

Here $G^{R,A}(\omega,\mathbf{k})$ and $\Xi(\mathbf{k},\mathbf{k}_0)$ are time integration of (9) and (10) between $\tau=0$ and ∞. Both the retarded and advanced Green's functions G^R and G^A are evaluated in the Born approximation for the scattering effects as

$$G^{R,A}(\omega,\mathbf{k}) = \frac{1}{\omega-\omega(\mathbf{k})\pm i(\gamma_0+\gamma)}, \tag{11a}$$

where $\gamma = \pi N(\omega)n_i|V_0(0)|^2$, with n_i the density of scattering centers, $N(\omega)$ the state density of the EP at ω, and $2\gamma_0$ the inelastic scattering rate, i.e., sum of exciton decay rate and acoustic phonon scattering rate of the EP. We evaluated $\Xi(\mathbf{k},\mathbf{k}_0)$ following the procedure of (16) of Ref. 10. Then we have

$$\Xi(\mathbf{k},\mathbf{k}_0) = \frac{-1}{2\gamma_0(\gamma_0+\gamma)^2}(\Gamma_c+\Gamma_l), \tag{11b}$$

where Γ_c and Γ_l come from summation of the maximally crossed diagrams and the ladder diagrams, respectively. Here

$$\Gamma_c(\mathbf{k}_0+\mathbf{k}) = \frac{2(\gamma_0+\gamma)U_0}{2\gamma_0+D(\mathbf{k}+\mathbf{k}_0)^2}, \tag{11c}$$

$$\Gamma_l(\mathbf{k}_0-\mathbf{k}_0') = \frac{2(\gamma_0+\gamma)U_0}{2\gamma_0+D(\mathbf{k}_0-\mathbf{k}_0')^2}, \tag{11d}$$

where in the ladder diagram we assumed two incident beams $\mathbf{k}_0$ and $\mathbf{k}_0'$ but we should put $\mathbf{k}_0'=\mathbf{k}_0$ for a case of single beam pumping. Here $U_0 = n_i|V(0)|^2$ and the diffusion constant $D = \nu_g^2/6(\gamma_0+\gamma)$ with the group velocity ν_g of the two-dimensional (2D) EP. Then we have finally the expression for the signal intensity as follows:

$$I_s = \frac{4A(\gamma_0+\gamma)}{[\omega_s+\omega_d-\omega_0-\omega(\mathbf{k})]^2+(\gamma_0+\gamma)^2}\frac{U_0}{\gamma_0^2(\gamma_0+\gamma)^2} \times\left[1+\frac{2\gamma_0}{2\gamma_0+D(\mathbf{k}+\mathbf{k}_0)^2}\right] \tag{12a}$$

$$\approx\frac{4AU_0}{\gamma_0^2(\gamma_0+\gamma)^2}\left[1+\frac{1}{1+D(\mathbf{k}+\mathbf{k}_0)^2/2\gamma_0}\right]. \tag{12b}$$

From this result and (6), it is clear that the signal intensity I_s is proportional to $|\alpha|^4$, and is thus proportional to the square of the incident power I_0 as observed by Rhee *et al.*[15] Here the first and second terms in the square brackets in (12) come from the ladder diagrams (11d) and the maximally crossed diagrams (11c), respectively. The difference of wave vectors on the left- and right-hand sides in Fig. 1 is conserved in the ladder-type scattering and it results in a uniform distribution of polaritons, since the rate (11d) is independent of $\mathbf{k}$ and $\mathbf{k}_0'=\mathbf{k}_0$ for the single-beam pumping. The maximally crossed diagrams result in an enhancement of the scattering for $\mathbf{k}=-\mathbf{k}_0$, and the backscattering is enhanced for $\mathbf{k}=-\mathbf{k}_0$ as (11c) shows. In the regime where the signal is proportional to the square of the pump power, the signal in the normal direction is four times as strong as in other directions. The factor 2 comes from the enhancement of backward scattering as shown in the square bracket in (12) and another factor 2 comes from the fact that both the signal and idler become coincident for the emission in the direction normal to the well. The angle of enhancement is within $\theta_c \equiv 2\gamma_0/Dk_0^2 = 12\gamma_0(\gamma_0+\gamma)/(k_0\nu_g)^2$. Because the EP suffers only from elastic scatterings the signal light has the same frequency and phase as the incident light within a time $1/\gamma_0$, and is thus coherent with the pump. For times longer than $1/\gamma_0$, the EP suffers from inelastic scattering, so the interference persists for a time $1/\gamma_0$ consistent with the observation of Rhee *et al.*[15]

The wave nature of the exciton may be manifested in the following way. A Gaussian pump beam is incident at a nonzero angle on the microcavity, creating exciton polaritons with wave vector $\mathbf{k}_0$. The exciton component of the EP has an initial central wave vector $\mathbf{k}_0$ and a spread in $\mathbf{k}$ space of width $\Delta\mathbf{k}$ which corresponds to the spread in spatial frequencies of the incident pump beam. The coherent backscattering generates a wave with central wave vector $-\mathbf{k}_0$. The angular spread of the backscattered wave is somewhat larger than the initial spread $\Delta\mathbf{k}$ due to the nonzero angular width of the coherent backscattering. If the divergence of the incident Gaussian beam is larger than the backscattering width, then the backscattered wave will have a spread close to $\Delta\mathbf{k}$. When the incident and backscattered exciton waves collide, a coherent polarization is created at $\mathbf{k}=\mathbf{0}$ with an angular width also close to $\Delta\mathbf{k}$, so that the emission into the normal direction has nearly the same angular spread as the incident pump beam (this can be narrower than the angular spread defined by the cavity[15]).

This wave model for the generation of the normal emission may be contrasted to the prediction of a particle model for the exciton collision. In that case, collisions of excitons with wave vector $\mathbf{k}_0$ and $-\mathbf{k}_0$ may result in a broad spread in final momenta, due to the random distribution of impact parameters for the two-particle collision. Thus a particle model would predict that at low density, the normal emission angular spread would be defined by the cavity. At very low density, in the linear response regime, the coherent backscattering intensity is expected to be twice the background random scatter intensity, as is typical for linear backscattering, and no special emission peak in the normal direction occurs. As the density is increased, however, the exciton-exciton collision rate increases and the $\mathbf{k}=0$ population (and the normal-direction emission) increases as square of the pump intensity.

Now we consider the effect of the microcavity. We suppose that the exciton and cavity modes are exactly resonant at $\mathbf{k}=\mathbf{0}$. Then the photon density of states is maximum at $\mathbf{k}=\mathbf{0}$, and $\mathbf{k}=\mathbf{0}$ EP is most efficiently coupled to the external field; we define the number of EP at $\mathbf{k}=\mathbf{0}$ to be n. The largest population of EP in k space is in the initially excited $\mathbf{k}=\mathbf{k}_0$ region. The coherent backscattering results in a population at $\mathbf{k}=-\mathbf{k}_0$ occurring with a rate given by (11c). The $\mathbf{k}_0$ and $-\mathbf{k}_0$ EP's collide [via the H'' term in (3c)] to produce a population at $\mathbf{k}=\mathbf{0}$. Further increase of the density results in stimulated emission into the $\mathbf{k}=\mathbf{0}$ state, as expressed by the $(n+1)(n+2)$ bosonic enhancement of the scattering rate. In this case, the $\mathbf{k}=\mathbf{0}$ population increases rapidly at the expense of the backscattered wave. This may be thought of as a condensation into the $\mathbf{k}=\mathbf{0}$ state, resulting in superradiance into the normal direction. This is because the population at $\mathbf{k}=\mathbf{0}$ increases due to the stimulated scattering (3c) but the emission is not the stimulated one as the decay rate of the $\mathbf{k}=\mathbf{0}$ EP is so rapid because of low cavity Q value. The angular spread of this emission is essentially the same as the incident pump divergence Δk at low density, and is reduced at high density by the stimulated scattering into exactly $\mathbf{k}=\mathbf{0}$.

The following aspects are predicted from this model. If the inelastic scattering rate is determined mainly by the acoustic-phonon scattering (i.e., the cavity radiative decay rate is smaller than the phonon scattering rate), then the signal I_s is expected to be inversely proportional to the square of the lattice temperature when $\gamma \gg \gamma_0$ as the signal intensity is inversely proportional to γ_0^2. Second, I_s is also proportional to $n_i/\{(\gamma_0+\gamma)^3\gamma_0^2\}$ so that there exists an optimum concentration of elastic scatters at which the signal shows maximum with temperature and pump power fixed.

The signal polariton ω_s comes from the polariton with the 2D wave vector almost equal to zero. When EP's backward-scattered into $\mathbf{k}=-\mathbf{k}_0$ are highly populated, both the signal ω_s and the idler ω_d have the zero vector for the 2D EP, and contribute to the signal. Therefore for such a case as the maximally crossed diagrams contribute, i.e., $|\mathbf{k}+\mathbf{k}_0|^2 \ll 2\gamma_0/D = 12\gamma_0(\gamma_0+\gamma)/\nu_g^2$ the observed signal increases by four in comparison to other directions even at low density. At high density, the normal emission will become dominant due to the stimulated scattering into the $\mathbf{k}=\mathbf{0}$ mode over the diffuse scattering. The EP's with zero and finite in-plane wave vector transmit very easily in the microcavity with the distributed Bragg reflectors on both ends. Therefore, we may expect two-step structures of the signal reflecting the distribution of the EP's over the in-plane wave vector $\mathbf{k}$ as a function of emitted angle from the normal direction. Some of these observations will give us further support of the weak localization of the EP.

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