Физика твердого тела, 1998, том 40, № 5

# Weak localization and nonlinear optical responses of exciton polaritons

© Eiichi Hanamura

Department of Applied Physics, University of Tokyo,
7–3–1 Hongo, Bunkyo-ku, Tokyo 113, Japan

We discuss novel nonlinear optical responses due to weak localization of exciton-polaritons (EP's): first phase-conjugated signal induced and enhanced by the weak localization of EP's under a single pump and a test beam which are two-photon-resonant to a biexciton. This observation will give us clear evidence of the weak localization. Second, we analyze, in terms of weak localization of EP's, the coherent light emission in the direction normal to the surface under a single beam pumping tilted from the normal to a planar semiconductor microcavity.

Since phase-conjugated wave (PCW) generation was shown theoretically to be enhanced by weak localization of exciton or excitonic polaritons (EP's) [1], several theoretical papers [2–5] were published for the effects of weak localization on optical nonlinearities. No experimental observation, however, of these effects has been reported yet. For the case of the conventional phase-conjugation process, three beams are necessary, and forward and backward pump beams are required to irradiate the front and rear surfaces at the frequency with a finite absorption coefficient. This configuration makes these experiments difficult. On the other hand, we have analyzed, in terms of the weak localization of EP's, coherent light emission in the direction normal to the surface under the tilted pumping in a planar semiconductor microcavity containing quantum wells (QW's) [6,7].

We propose first in the present work how to observe much clearer evidence of weak localization of the excitons of EP's under a single pump and a test beam irradiating a crystal surface and nearly two-photon resonant to a biexciton [8]. The main result is as follows: Let us denote by $\theta$ the angle between the pump and test beams. Then it will be shown that for $\theta$ larger than the critical angle $\theta_c$, the phase-conjugated wave is induced and enhanced by the weak localization in the direction inverse to the test beam but that no clear enhanced signal is expected for the case $\theta < \theta_c$. The expression of $\theta_c$ will be given later. In the latter case, the sharp signal due to the weak localization disappears so that its observation as a function of $\theta$ gives us its direct evidence.

The EP excited by the forward pump wave is dominantly scattered into the backward direction due to the effect of weak localization of the EP. Here the backward-scattered EP plays the same role as the EP created by the backward pump wave. Without the weak localization, the PCW is missing. Therefore the observation of PCW gives direct evidence of weak localization. This weak localization comes from constructive interference of multiple scattering of the EP and its time reversed process. Multiple scatterings of the EP are composed of the two diagrams, i. e., the ladder-type diagram $\Gamma_l$, and the maximally crossed diagram $\Gamma_c$, respectively. These effects are summed up into the following forms [1]:

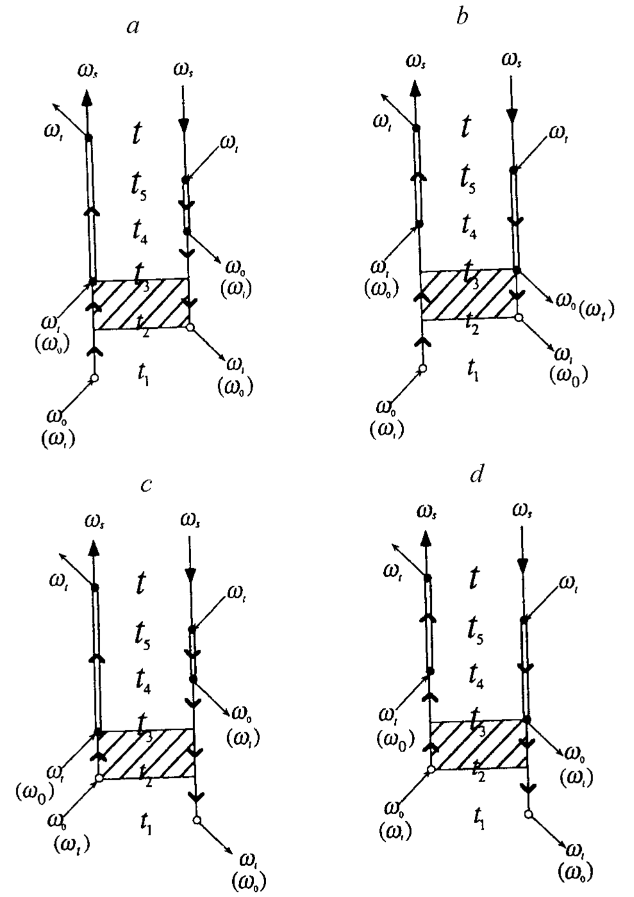
$$\Gamma_l\big(\mathbf{k}_t - \mathbf{k}_0\big) = \frac{2(\gamma_0+\gamma)U_0}{2\gamma_0 + D(\mathbf{k}_t - \mathbf{k}_0)^2}, \qquad (1)$$

$$\Gamma_c\big(\mathbf{k}_t + \mathbf{k}_0\big) = \frac{2(\gamma_0+\gamma)U_0}{2\gamma_0 + D(\mathbf{k}_t + \mathbf{k})^2}. \qquad (2)$$

Here $\mathbf{k}_t$, $\mathbf{k}_0$ and $\mathbf{k}$ are wave vectors, respectively, of EP's excited by the test and pump beams and the signal beam, $\gamma_0$ is the inelastic scattering rate of the EP, $\gamma \equiv \pi N(\omega) n_i |V(0)|^2$ with the state density $N(\omega)$ of the EP, $n_i$ is the number density of elastic scatterers and $V(0)$ is the zero-Fourier component of the potential due to the elastic scatterer, $U_0 \equiv n_i|V(0)|^2$ and $D \equiv v_g^2/6(\gamma_0+\gamma)$ is the diffusion constant of the EP with its group velocity $v_g$.

For the case $\theta < \theta_c$, $|\mathbf{k}_t - \mathbf{k}_0|^2/\mathbf{k}_0^2 \approx \theta^2 < \theta_c^2 \equiv 2\gamma_0/D\mathbf{k}_0^2$ so that we may put $\mathbf{k}_0 = \mathbf{k}_t$ in (1). Then the ladder-type contribution $\Gamma_l$ gives the stronger scattering of the EP into any directions. On the other hand, under such a two-beam pumping as $D(\mathbf{k}_t - \mathbf{k}_0)^2 > 2\gamma_0$, i. e. $\theta > \theta_c$, the ladder-type scatterings $\Gamma_l$ are negligible in comparison to the maximally crossed ones with $\mathbf{k} \approx -\mathbf{k}_t$. As a result, the backward scattering of the EP excited by the test beam is most effectively brought about and the phase-conjugated signal is observable for $\theta > \theta_c$. While the signal intensity of conventional phase-conjugated wave is proportional to $I_p^2 I_t$, the present signal has the $I_p I_t^2$ dependence. This is understood from the diagram of Fig. 1, where two-photon resonance of biexciton ($\omega_s + \omega_t = \omega_b$: the biexciton frequency) is made of use so that the biexction-photon interaction is used twice and the EP-photon one is once [8]. Here $I_t(I_p)$ and $\omega_t(\omega_p)$ are the intensity and frequency, respectively, of the test (pump) beam. Another interesting point of this process is that when the biexciton is resonantly excited by the two-photon process $\omega_s + \omega_t = \omega_b$, the phase-conjugated signal is enhanced not only by the resonance effect but also by the giant oscillator strength for the transition between the states of a signal exciton and a biexciton [8,9]. As a conslusion, the observation of PCW as a function of the angle between pump and test beams will give as a clearer evidence of weak localization.

Second, we will describe the observation by Rhee et al. on the coherence transfer between EP's in a system consisting of multiple-quantum-well excitons resonantly coupled to a planar Fabry–Pérot microcavity [6]. Specifically, they observed the emission of light in the direction perpendicular to the surface with a small divergence angle, even though the 2D EP states were excited at a much larger angle (3°) away from the normal. The emitted light was confirmed to be coherent with the incident pump light by interfering the emission with the pump beam. The emitted light intensity was also observed to increase more strongly than by the second power of pump light intensity. These experimental




Diagrams contributing to the generation of phase-conjugated exciton polariton $\omega_s$. Solid line and double solid line describe propagation of an EP and a biexciton, respectively, and thin lines describe the external fields of pump $\omega_0$ and test $\omega_t$ beams.

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 mainly into states with momentum $-\mathbf{k}$. Collisions between EP's with $\mathbf{k}_0$ and $-\mathbf{k}_0$ result in generation of a population of EP's at exactly $\mathbf{k} = 0$, giving rise to coherent emission of light in the normal direction. Note here that the bosonic character is reflected in the scattering and collisional processes, i. e., EP's are most strongly scattered into the most highly populated states $-\mathbf{k}_0$ and $\mathbf{k} = 0$.

We assume that the incident laser tilted at an angle $\theta$ from the normal excites a coherent state $|\alpha\rangle$ of the EP with an in-plane momentum $\mathbf{k}_0$ in a well. The emission intensity in the normal direction may be considered to be proportional to the number of EP's generated in the single mode per second, and may be evaluated under CW excitation as

$$I_s = \lim_{t\to\infty} \frac{\partial}{\partial t} \langle\langle c_0^+ c_0 \rho(t) \rangle\rangle. \tag{3}$$

The double angular brackets in (3) signify both the quantum-mechanical and ensemble average over the distribution of scattering potentials. An irrelevant factor, coming from transmissivity determined by coupling of the EP's at $\mathbf{k} = 0$ to the external electric field, has been omitted. The density operator $\rho(t)$ of the total system is

$$\rho(t) = \exp(-iH_T t)\, \rho_0 \exp(iH_T t), \tag{4}$$

in terms of the total Hamiltonial $H_T$. We expand (4) in $H'$ the interaction of EP's with the external electromagnetic field $\mathbf{E}_j$. The lowest-order contribution comes from the fourth-order term, i. e. second-order in the pump and second-order in the signal field. According to the calculations of [1,3], (3) is evaluated by taking account of multiple scattering of EP by the elastic scattering potential $V_0(\mathbf{q})$, i. e., the effect of maximally crossed $\Gamma_c$ and ladder-type $\Gamma_l$ diagrams. Then $I_s$ expressed in the lowest approximation in external fields:

$$I_s = \frac{4AU_0}{\gamma_0^2(\gamma_0+\gamma)} \left[ 1 + \frac{1}{1 + D(\mathbf{k}+\mathbf{k}_0)^2/2\gamma_0} \right], \tag{5}$$

$$A = \left| \left\langle n_0+1, \alpha \middle| c_0^+ c_0^+ H'' \middle| n_0, \alpha \right\rangle \left\langle \alpha \middle| c_\alpha^+ \boldsymbol{\mu} \cdot \mathbf{E} \middle| \alpha \right\rangle \right|^2. \tag{6}$$

Here $\gamma_0$ is the inelastic scattering rate, $\gamma \equiv \pi N(\omega) n_i |V_0(0)|^2$ with the EP state density $N(\omega)$ and the number density of scatterers $n_i$ , $U_0 \equiv n_i |V_0(0)|^2$, $D \equiv v_g^2/6(\gamma_0+\gamma)$ is the diffusion constant of EP with its group velocity $v_g$ and the transition dipole moment $\boldsymbol{\mu}$ created by $c_\alpha^+ \equiv c^+(\mathbf{k}_0)$. From (5) and (6), the signal intensity is proportional to the square of the incident power $I_0$ in the weak pumping and increases more strongly against $I_0$. This is because the induced scatterings into $\mathbf{k} = 0$ and $-\mathbf{k}$ work, respectively, in the EP–EP collision Hamiltonian $H''$ and $V_0(\mathbf{q})$ term. There the sharp peak in the normal direction is understood as a combined effect of the weak localization and induced scattering to this mode, which has the longest life-time in the semiconductor microcavity. The angle of enhancement is within $\theta_c \equiv 2\gamma_0/Dk_0^2 = 12\gamma_0(\gamma_0+\gamma)/(k_0 v_g)^2$. Because the EP suffers only from elastic scatterings, the signal light has the same frequency and phase as the incident light within a lifetime $1/2\gamma_0$ and is thus coherent with the pump beam. For times longer than $1/\gamma_0$, the EP suffers from inelastic scattering, so that the interference persists for a time $1/\gamma_0$. These results are consistent with the observation of Rhee et al. [6].

## References

[1] E. Hanamura. Phys. Rev. **B39**, 1152 (1989).
[2] V.E. Krabtsov, V.I. Yudson, V.M. Agranovich. Phys. Rev. **B41**, 2794 (1990).
[3] A.R. McGurn, T.A. Leskova, V.M. Agranovich. Phys. Rev. **B44**, 11 441 (1991).
[4] V.I. Yudson, P. Reineker. Phys. Rev. **B45**, 12 470 (1993).
[5] N. Taniguchi, E. Hanamura. Phys. Rev. **B47**, 2073 (1993).
[6] J.K. Rhee et al. In: Ultrafast Phenomena. OSA (1996). P. 220.
[7] E. Hanamura, T.B. Norris. Phys. Rev. **B54**, R2292 (1996).
[8] E. Hanamura. Phys. Rev. **B54**, 11 219 (1996).
[9] E. Hanamura. Solid State Commun. **12**, 951 (1973).