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Journal: Physical Review B
Volume: 63
Number: 23
Page range: 233203
Year: 2001-06

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URL: http://hdl.handle.net/2241/98308
doi: 10.1103/PhysRevB.63.233203
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(Received 28 February 2001; published 30 May 2001)

We report observation of the inverse polariton series and a detailed experimental study of the exciton components in polaritons (Hopfield coefficients). Spontaneous emission of excitonic molecules into outgoing polaritons associated with the \( i = 1,2,3,4,5 \)th exciton states in bulk CuCl is detected and analyzed by the biexciton model. Because the intensities of the emission lines, which form the inverse polariton series, are determined by the exciton components in the final-state polaritons, we are able to measure the Hopfield coefficients for the highly composite outgoing polaritons. Quantum interference in the optical decay of excitonic molecules, due to the multiple exciton components in the polariton and biexciton states, is also demonstrated.

DOI: 10.1103/PhysRevB.63.233203 PACS number: 71.35.—y, 71.36.+c, 78.55.Hx, 42.65.Yj

The quantum theory of light that resonates with excitons was developed by Hopfield in 1958.1 The Hamiltonian for the coupled polarization and electromagnetic fields can readily be diagonalized to generate new eigenstates, polaritons. The polariton eigenstates are characterized by the polariton dispersion and the Hopfield coefficients. The latter describe the relative contributions of the photon and exciton components to the polariton state. The Hopfield coefficients naturally arise in the theoretical descriptions of many phenomena such as polariton scattering,2–6 polariton squeezing,7,8 and biexciton radiative decay.9,10 The polaritons associated with the optically active ground-state excitons in bulk semiconductors have been extensively studied in numerous experiments.11 Most of the experiments, however, deal only with the polariton dispersion, while so far there have been no clear measurements of the Hopfield coefficients, for the following reasons. First, in Raman/Brillouin scattering or four-wave mixing experiments,12 the Hopfield coefficients combine with the corresponding polariton density of states so that the relevant nonlinear susceptibilities are insensitive to the Hopfield coefficients.3,4,13 Secondly, for polaritons in bulk direct-band-gap semiconductors, the Rabi splitting energy is usually larger than the energy band that contributes to the optical signal, i.e., the excitonlike limit of the polariton is adequate in the cases so far studied.1,5 By contrast, for microcavity polaritons,5,14 the Rabi splitting and the density of states are controlled artificially. Thus the Hopfield coefficients can indeed be experimentally visualized as proposed in Ref. 15, although a rigorous comparison of experiments with theories is absent due to the structural complexities of such systems. In addition to cavity polaritons, a wide variety of artificial polariton systems have been realized in recent years. For example, coupled microcavities and periodic structures of quantum wells and dots16 have been fabricated, leading to a clear demonstration of polariton coupling. These exhibit intrinsically multiple-band polaritons, thus requiring the Hopfield theory to be generalized to multiple oscillators.

In this Brief Report, by studying the optical decay of excitonic molecules (biexcitons) into polaritons associated with the \( ns \) exciton, we report a detailed experimental analysis of the Hopfield coefficients in bulk CuCl. In the presence of exciton-exciton interaction, the polariton normal modes are no longer independent oscillators. In the extreme case of biexciton formation the perturbative approach is not valid either for exciton-exciton interaction or for exciton-photon coupling. In this case, in order to treat the above interactions nonperturbatively, one should apply the bipolariton model of an excitonic molecule.17 For the resonant dissociation of the biexciton \( K_m \) into two outgoing polaritons \( k_1^{\text{out}} \) and \( k_2^{\text{out}} \), the intensity of the optical signal associated with the lower-branch polariton (\( \mu = 1 \) \( k_1^{\text{out}} \)) is given by

\[
I_{\mu, i}(k_1^{\text{out}}) \approx \rho^{(1,2)} \left| B_{1s}^{(\mu=1)}(k_1^{\text{out}}) B_{1s}^{(\nu)}(k_2^{\text{out}}) \right| e^m \int dR |\Psi_m(R)|^2,
\]

where \( \rho^{(1,2)} \) is the joint density of the polariton states, \( e^m \) is the binding energy of the molecule, \( B^{(\nu)}(k) \) is the Hopfield coefficient that characterizes the amplitude of the excitonic polarization associated with the \( i \) state in the \( \nu \)-branch polariton, and \( \Psi_m(R) \) is the biexciton envelope wave function (WF) as a function of the coordinate \( R \) of the relative motion of the two constituent excitons. Note that the efficiency of the spontaneous decay of biexcitons given by Eq. (1) explicitly depends on the Hopfield coefficients \( B^{(\nu)}(k) \), thus providing us with a unique possibility to measure the Hopfield coefficients.

Single-crystal CuCl is a prototype material in the physics of polaritons and biexcitons because of the relatively simple valence band structure, the large binding energies of the ground-state exciton and biexciton (\( \epsilon^{(V,1)} \approx 200 \text{ meV} \) and \( \epsilon^{m} \approx 32 \text{ meV} \), and the large polariton Rabi frequency (\( \hbar \Omega_{x(1)} \approx 224 \text{ meV} \)). Since the Rabi frequency (the energy scale for the polariton effect) is larger than the biexciton binding energy, the bipolariton model17 is necessary to describe the optical decay of molecules. As shown in Fig. 1, below the band gap (\( \approx 3.4 \text{ eV} \)) there are \( Z_3 \) 1s, \( Z_{1,2} \) 1s,18 and \( Z_2 \) 2s, 3s, and 4s excitons.19 These states give rise to the five polariton branches LPB, UPB, and PB2,3,4, re-
spectively. In Fig. 1 we show a graphical solution of the energy-momentum conservation law in the optical decay of the biexciton state $K_m = k_1^{\text{in}} + k_2^{\text{in}}$ into multi-
branch polaritons $k_1^{\text{out}} + k_2^{\text{out}}$ and $k_2^{\text{out}} + k_2^{\text{out}}$. The outgoing polaritons refer to the crossing points between the plotted dispersion curves (in our experiments the signal lower-branch polariton $k_1^{\text{out}}$ is detected in the forward direction, while the conjugated backward-scattered polariton $k_2^{\text{out}}$ is idle). If the biexciton resonant decay into the PB$_n$ ($n > 1$) branches is observed, the analysis in the excitation energy limit is no longer adequate because of a wider energy range of observation than the polariton Rabi splitting.

Two synchronized tunable uv picosecond pulses of 2 ps duration, 82 MHz repetition, and 20 mW average power are delivered through doubling crystals from two TiS lasers electrically synchronized by a Lok-to-Clock circuit (Spectra Physics). The high-repetition-rate picosecond pulses are important for high spectral resolution and for avoiding the higher-order nonlinearities due to reexcitation of biexcitons. For two-photon generation of biexcitons in CuCl, we change the laser frequencies $\omega_1^{\text{in}}$ and $\omega_2^{\text{in}}$, keeping the sum of them resonant with the biexciton energy, i.e., $h \omega_1^{\text{in}} + h \omega_2^{\text{in}} = h \Omega_m = 6.372$ eV. The two circularly polarized laser beams, which are aligned antiparallel and focused into a high-quality CuCl single crystal at 2 K, induce counterpropagating lower-branch polaritons with wave vectors $k_1^{\text{out}}(\omega_1^{\text{in}})$ and $k_2^{\text{out}}(\omega_2^{\text{in}})$. The incoming polaritons selectively generate cold molecules with $K_m = k_1^{\text{in}} + k_2^{\text{in}}$ which spontaneously decay into outgoing polaritons $k_1^{\text{out}}$ and $k_2^{\text{out}}$, mainly before the incoherent scattering processes occur. Emitted light due to the outgoing polaritons is detected by a Si charge coupled device camera through a standard fiber bundle and a monochromator.

The emission spectra collected from the transient biexciton states from the band $0 \leq |K_m| \leq 0.6k_0$ are plotted in Fig. 2. $[K_m = 2k_0$ is the biexciton wave vector of degenerate $(k_1^{\text{in}} = k_2^{\text{in}} = k_0)$ two-polariton excitation, $k_0 = 0.44 \times 10^6$ cm$^{-1}$].

The spontaneous decay $K_m - k_1^{\text{out}}(LPB) + k_2^{\text{out}}(UPB)$ gives rise to the doublet of spectral lines called LP and UP. In Fig. 2, as well as the LP signal, very weak replicas labeled LP$_{2,3,4}$ are also seen on the lower-energy side, due to the channels $K_m - k_1^{\text{out}}(LPB) + k_2^{\text{out}}(PB_{2,3,4})$. The LP and UP lines form the inverse polariton series. The weak (the intensity ratios $I_{LP_{2,3,4}}/I_{LP}$ are of the order of 1/100) but finite transition probabilities into PB$_{2,3,4}$ ($n > 1$) arise from the 1$s$ exciton components in the outgoing polaritons.

The experimentally determined multibranch polariton dispersion and the component of the $Z_3$ 1$s$ exciton in the polariton state are shown in Figs. 3(a) and 3(b) by the filled circles. The solid curves are calculated by a five-oscillator potential model described below. Figure 3(b) clearly shows how strong is the exciton-photon interaction in CuCl; it re-
distributes the $Z_3$ 1$s$ exciton component $B_{1,2}$ in the entire energy region over the five-branch polariton dispersion.

The polariton dispersion associated with $N$ exciton states, each characterized by the principal quantum number $n$ and zero orbital angular momentum, is given by

$$\frac{e^2 k^2}{\hbar \varepsilon_b \omega^2} = 1 + \sum_{n=1}^{N} \frac{\Omega_{X(n)}^2}{\omega_{X(n)}^2 - \omega^2}, \quad (2)$$

where $\varepsilon_b$ is the background dielectric constant, $\hbar \omega_{X(n)}$ is the energy of the $X(n)$ exciton state, and $\Omega_{X(n)}$ is the corresponding polariton Rabi frequency. The solid curves in Fig. 3 are given by Eq. (2) with $\varepsilon_b = 4.3$, $\hbar \omega_{Z_3(1s)} = 3.2022$ eV, $\hbar \omega_{Z_3(1s)R} = 3.2670$ eV, $\hbar \omega_{Z_3(2s)} = 3.3665$ eV,
The subject of the document is about the analysis of experimental data and theoretical models related to the behavior of polaritons in solid-state systems. The text discusses the evaluation of experimental points in Fig. 3, the determination of the best fit to the data, and the theoretical curves calculated by Eqs. (2)–(4) that agree well with the experimental data. The document highlights the importance of residual differences between experimental and theoretical results and the consistency of the theoretical curve with the experimental points. Additionally, it notes the occurrence of some discrepancy between the experimental and theoretical results in the spectral region close to the (2,3) s exciton resonances. The text also mentions that the theoretical curve is consistent with the deviation of the binding energies from the 1/n^2 law, and it suggests that in order to fit the dispersion, the oscillator strengths are found to be less than one-half of those estimated by the 1/n^3 law for n = 2,3,4. Furthermore, the document indicates that for B_{1s}^{(3,4)} plotted in Fig. 3(b), there is some discrepancy between the experimental and theoretical results in the spectral region close to the (2,3) s exciton resonances.
The two spontaneous decay paths interfere constructively to give nearly equal contributions to the LP$_2$, intensity at $K_m = 0$, which should otherwise be much less than observed. Further, the LP$_3_s$ line is determined by three paths (1,1), (1,2), and (1,3), i.e.,

$$ I_{LP_3} \propto |B_{1s}^{(1)}(k_1^u)B_{2s}^{(4)}(k_2^u)W_{1,1}C_{1,1} + B_{1s}^{(1)}(k_1^u)B_{2s}^{(4)}(k_2^u)W_{1,2}C_{1,2} + B_{1s}^{(1)}(k_1^u)B_{2s}^{(4)}(k_2^u)W_{1,3}C_{1,3}|^2. \tag{7} $$

For $|K_m| = 0$ the first and third paths are dominant and interfere constructively. For $K_m > 0.05 \times 10^6$ cm$^{-1}$, the LP$_3_s$ line almost disappear (see Fig. 2) due to destructive interference of the second path with the first and third ones. The $K_m$ dependence of the LP$_3_s$ line is explained similarly.

In conclusion, we have measured the Hopfield coefficients for highly composite polaritons in CuCl and verified the Hopfield theory generalized to multioscillator polaritons.

We appreciate valuable discussions with M. Kuwata-Gonokami and N. Nagasawa, and thank K. Kurihara for sample purification. A.L.I. acknowledges support by EPSRC.