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Exciton–biexciton quantum coherence and polaritonic stop-band transparency in CuCl

S. Chesi[1], **M. Artoni**[2], **G. C. La Rocca**[*, 3], **F. Bassani**[3], and **A. Mysyrowicz**[4]

[1] Dept. of Physics, Purdue University, West Lafayette, IN 47907-1396, USA
[2] Dipartimento di Chimica e Fisica per l'Ingegneria e per i Materiali, Università di Brescia, Via Valotti 9, 25133 Brescia, Italy
[3] Scuola Normale Superiore and INFM, Piazza dei Cavalieri 7, 56126 Pisa, Italy
[4] Laboratoire d'Optique Appliquée, ENSTA, École Polytechnique, 91761 Palaiseau Cedex, France

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A coherently driven exciton–biexciton transition in CuCl enables one to propagate a probe light beam within the exciton-polariton stop-band where radiation is otherwise completely reflected. The stop-band transparency window can be controlled via the pump beam frequency and intensity. The phenomenon is reminiscent of quantum coherence effects occurring in three-level atomic systems, except that it here involves delocalized electronic excitations in a crystal via a frequency and wave-vector selective polaritonic mechanism. Both a free standing slab and a microcavity configuration are theoretically studied.

Phenomena based on three-level quantum coherence in atomic physics have been of considerable importance in recent years. Ranging from rather familiar effects, such as electromagnetically induced transparency (EIT) and lasing without inversion (LVI), to currently developing applications of ultraslow light, these phenomena all depend on the existence of quantum–coherence in a multi–level system [1, 2]. We here report on effects of quantum coherence between exciton and biexciton [3] levels in CuCl. CuCl is a prototype example of a semiconductor having an allowed interband transition, quite pronounced exciton and biexciton resonances [4] and exhibiting, in particular, a fully developed polaritonic stop-band. We predict that a pump driven exciton−biexciton transition allows for a well developed transparency within the stop-band where a probe pulse may propagate, although with a strongly reduced group velocity [5]. More specifically, we show how the transparency of a probe beam within the Z_3-exciton polariton stop-band can be controlled via a pump light beam resonant with the transition from the Z_3-exciton to the Γ_1-biexciton. The large oscillator strength of the exciton–biexciton transition and the very narrow linewidth and long coherence time of the biexciton state in the small wave-vector region [6, 7] appear to favor quite appreciable degrees of transparency. The phenomenon is reminiscent of EIT effects occurring in three-level atomic systems [1, 8], except that in CuCl delocalized electronic excitations in a crystalline structure are involved instead [9]. Unlike in atomic-like media, the physics of the induced transparency within an otherwise reflecting stop-band relies on a frequency and wave-vector selective polaritonic mechanism.

Transparency can be induced in our case through a ladder scheme in which a circularly polarized probe beam is nearly resonant with the transition from the crystal ground state to the Z_3 exciton with dispersion $\omega_x(k) = \omega_T + \hbar k^2/(2m_x)$, while a pump beam having opposite circular polarization couples

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[*] Corresponding author: e-mail: larocca@sns.it

the Z_3 exciton to the Γ_1 biexciton with dispersion $\omega_m(k) = \omega_M + \hbar k^2/(2m_m)$. The CuCl response to a weak probe beam of frequency ω and wavevector $\boldsymbol{k}$, in the presence of the strong coupling beam of frequency ω_c and wave-vector $\boldsymbol{k}_c$ opposite to $\boldsymbol{k}$, turns out to be described by the following dressed dielectric constant [10, 11]

$$\varepsilon(k,\omega) = \varepsilon_b + \frac{\varepsilon_b \Delta_{\mathrm{LT}}}{\hbar\omega_x(k) - \hbar\omega - i\gamma_x + \Sigma} \quad \text{with} \quad \Sigma = \frac{\beta}{\hbar\omega + \hbar\omega_c - \hbar\omega_m(k - k_c) + i\gamma_m}. \tag{1}$$

Here Σ describes the nonlinearity due to the coherent pump and β is proportional to the pump intensity and the oscillator strength of the exciton–biexciton transition ($\beta \simeq 10^{-8}\,\mathrm{eV}^2$ at a pump power of 10 kW/cm^2). All the CuCl material parameters [5] appearing in Eq. (1), i.e., the exciton and biexciton $\boldsymbol{k} = 0$ energies $\hbar\omega_T$ and $\hbar\omega_M$, their masses m_x and m_m and linewidths γ_x and γ_m, the background dielectric constant ε_b, and the exciton longitudinal-transverse splitting Δ_{LT} are known from experiment. The exciton and biexciton linewidths have been introduced phenomenologically while the contribution of the Z_{12} exciton resonance at 3.267 eV [12] is included into the background dielectric constant ε_b. Small pump-induced nonresonant energy shifts [6] have been neglected as unimportant here. The main features of ε in Eq. (1) have also been confirmed by recent calculations at the $\chi^{(3)}$ level [3] that go beyond the partly phenomenological theory leading to Eq. (1).

When Maxwell's equations are solved with such an $\varepsilon(k,\omega)$ in the absence of the pump ($\beta \to 0$) one obtains the usual upper and lower polariton dispersion branches and a polaritonic stop-band within which the probe is nearly completely reflected [13]. In typical atomic systems, instead, the peak exciton absorption given by $\varepsilon_b\Delta_{\mathrm{LT}}/\gamma_x$ is about four orders of magnitude smaller than in the present case and, thus, the width of the polaritonic stop-band becomes negligible. The exciton–biexciton coherent coupling induced by the pump leads to three solutions of the probe dispersion equation $(c^2k^2/\omega^2) = \varepsilon(k,\omega)$ [14]. Thus, a third polariton branch appears in the frequency region of the exciton resonance, and this is expected to affect the probe reflectivity [6, 10, 15]. It is worth emphasizing that several polariton branches are present so that the knowledge of the local dielectric function ε is not sufficient to calculate the probe transmission through a coherently driven slab of CuCl. The solution of Maxwell's equations requires additional boundary conditions (ABC) [6, 13]. Here, we adopt the simplest ones, i.e., the extended Pekar's ABC [11] assuming both exciton and biexciton polarizations to vanish at the slab surfaces. This approach has been shown to agree with the results of a microscopic nonlocal response theory which does not require any ABC [11].

Our predictions show that transmission within the forbidden stop-band can indeed be completely controlled. We show in Fig. 1 the probe normal incidence transmission spectra through a CuCl slab 0.15 μm thick for different pump intensities and frequencies with the inclusion of the exciton and biexciton dampings. A pronounced transparency window in correspondence of the pump-induced dispersion branch opens up within the polaritonic stop-band around a frequency $\omega \simeq \omega_m(k - k_c) - \omega_c$. The transparency frequency can be coherently controlled over a rather wide spectral range of several meV corresponding to the entire polaritonic stop-band. However, we remark that even within the transparency window the absorption is here still quite significant. Absorption in a CuCl slab 0.15 μm

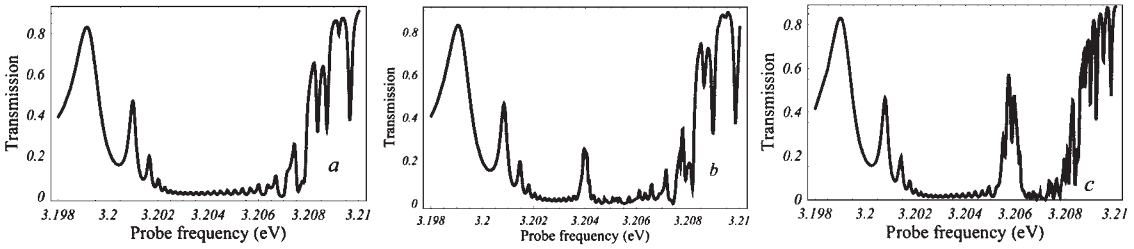

Fig. 1 Probe transmission spectra through a CuCl film 0.15 μm thick: a) no pump, b) with a pump of frequency $\hbar\omega_c = 3.168$ meV and intensity corresponding to $\beta = 5 \times 10^{-7}$ eV^2, c) with a pump of frequency $\hbar\omega_c = 3.166$ meV and $\beta = 10^{-6}$ eV^2. The exciton and biexciton linewidths are $\gamma_x = 50$ μeV and $\gamma_m = 15$ μeV.

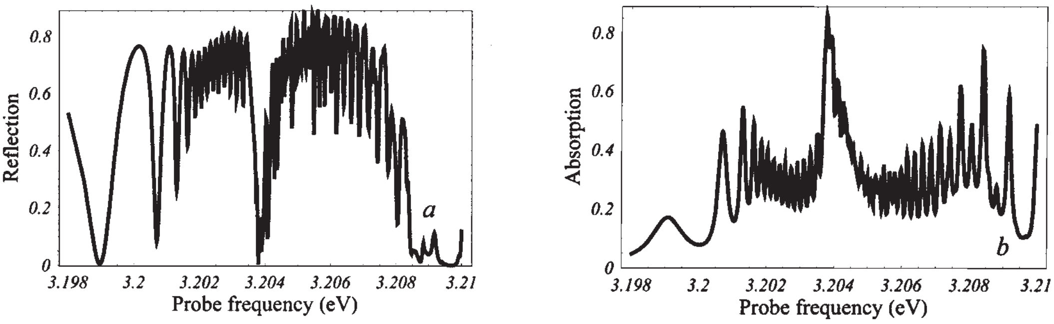

Fig. 2 Probe reflection a) and absorption b) spectra corresponding to the pump and material parameters as in Fig. 1b.

thick for a fixed pump intensity and probe frequency is shown in Fig. 2 along with the corresponding reflection spectrum. Unlike the exciton linewidth, the biexciton linewidth at a small wave-vector γ_m affects critically the appearance of a transparency window [5].

In strongly coupled semiconductor microcavities [16], the effects of biexciton resonances are a subject of current interest [17]. Also in these systems, we expect the exciton–biexciton coherent coupling to introduce in a similar way a third cavity polariton branch besides the usual upper and lower ones separated by the Rabi splitting. Using a material with well developed and strongly bound exciton and biexciton resonances such as CuCl would make all related effects quite visible. To the best of our knowledge, no experimental results have been reported on microcavities or quantum confined systems based on CuCl. In the following, we use model parameters extrapolated from the bulk values to simulate the behaviour of a microcavity containing a CuCl quantum well with no claim of being quantitative. The relevant physical scales are thus chosen as follows: the Rabi splitting is about 50 meV, the two-dimensional biexciton binding energy is about 100 meV, the two dimensional exciton and biexciton broadenings are of the order of 1 meV, the coupling parameter β is about 10^{-4} eV^2 [18]. The microcavity transmission and reflection spectra of the probe are obtained by a transfer matrix calculation in which the CuCl quantum well is included via a local dressed dielectric function of the same form as above Eq. (1). The cavity is tuned below the exciton resonance with a detuning close to the biexciton binding energy. The exciton–biexciton quantum coherence can then be driven by a pump resonantly entering the cavity at near normal incidence (pump and probe having opposite circular polarizations as above). In Fig. 3a the microcavity reflectivity dips are shown as a function of probe frequency and parallel wavevector q: the third cavity polariton branch induced by the pump is clearly visible. The corresponding cut for q close to the bare exciton-cavity mode anticrossing is shown in Fig. 3b, while Fig. 3c shows the respective absorption spectrum. In comparison to the free standing slab, it is to be noticed that much larger biexciton (and exciton) broadenings do not prevent

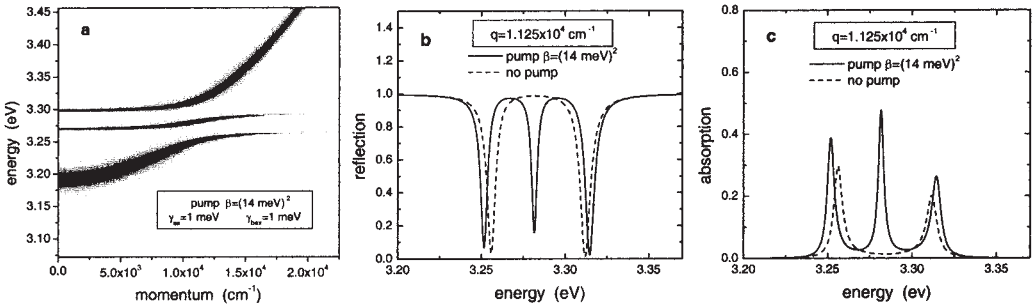

Fig. 3 a) Microcavity reflectivity dips as a function of probe frequency and parallel wavevector; b) reflectivity spectrum near the anticrossing; c) absorption spectrum near the anticrossing.

the observation of the pump induced transparency associated to the third cavity polariton branch. Finally, the exciton–biexciton coherent coupling effects here considered are of the same nature as those described by the "average polarization model" of Ref. [19], and quite distinct from effects related to the presence of an incoherent exciton population that transfers oscillation strength to the biexciton transition [20].

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