Polaritonic Stop-Band Transparency via Exciton-Biexciton Coupling in CuCl

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(Received 28 January 2003; published 29 July 2003)

Radiation is almost completely reflected within the exciton-polariton stop band of a semiconductor, as in the typical case of CuCl. We predict, however, that a coherently driven exciton-biexciton transition allows for the propagation of a probe light beam within the stop band. The phenomenon is reminiscent of electromagnetically induced transparency effects occurring in three-level atomic systems, except that it here involves delocalized electronic excitations in a crystalline structure via a frequency and wave-vector selective polaritonic mechanism. A well-developed transparency, favored by the narrow linewidth of the biexciton, is established within the stop band where a probe pulse may propagate with significant delays. The transparency window can be controlled via the pump beam detuning and intensity.

DOI: 10.1103/PhysRevLett.91.057402 PACS numbers: 78.20.–e, 42.50.–p, 42.65.–k, 71.36.+c

Electromagnetically induced transparency (EIT) in *atomic* media has received much attention in the past decade. The underlying physical mechanism, which relies on the destructive interference between several pathways connecting the atom's ground and excited states, is now well understood and has been the subject of several recent reviews [1]. EIT manifests itself in the formation of a transparency window within the atom's resonant absorption region accompanied by extremely slow-light propagation [2].

Most of the studies concern atomic systems and are based on typical three-level lambda or ladder configurations [1,2]. Theoretical studies of EIT in bulk semiconductors have considered the forbidden yellow exciton in Cu_2O in a lambda scheme involving the ground state and the intrinsic $1S$ and $2P$ exciton levels [3]. Furthermore, both excitonic and biexcitonic resonances [4] also exhibit a variety of three-level configurations where EIT could in principle be achieved, as recently suggested [5]. When comparing solid and atomic systems, important dissimilarities have to be taken into account. One is dephasing, typically orders of magnitude faster in solids than in atomic vapors, which can easily break the coherence of the population trapping state and which has made the observation of large EIT effects in solids rather difficult [6,7]. The other is the delocalized nature of the intrinsic exciton (biexciton) states having a well-defined wave vector $\vec{k}$ and a significant wave-vector dispersion [8], in strong contrast with the case of atomic levels having localized wave functions. Finally, an important difference consists in the polaritonic effects that typically occur for excitons with a large oscillator strength [9]. The Cu_2O exciton previously considered [3], however, has a weak oscillator strength and, hence, negligible polaritonic effects making the basic physics underlying the phenomenon of EIT in that case much similar to the one occurring in atoms.

In this Letter, we predict the possibility of observing EIT in CuCl which is a *non-atomic-like* crystal with a fully developed polaritonic stop band. This is a prototype example of a semiconductor having an allowed interband transition and pronounced exciton and biexciton resonances [10]. Specifically, we show how the transparency of a probe beam within the Z_3-exciton polariton stop band can be controlled via a coupling light beam resonant with the transition from the Z_3 exciton to the Γ_1 biexciton, i.e., the bound molecular state of two Z_3 excitons [10,11]. 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 [12,13] appear to favor quite appreciable degrees of transparency along with slow group velocity propagation regimes [14]. Unlike in atomiclike media, the physics of the reduced group velocity and 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

TABLE I. Material parameters of CuCl (low temperature values) [11,13,16,17].

$\hbar\omega_T$ (eV)	$\hbar\omega_M$ (eV)	m_x (m_o)	m_m (m_o)
3.2022	6.3720	2.30	5.29
γ_x (μeV)	γ_m (μeV)	ϵ_b	Δ_{LT} (meV)
50	15	5.59	5.65

opposite circular polarization couples 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 wave vector $\vec{k}$, in the presence of the strong coupling beam of frequency ω_c and wave vector $\vec{k}_c$ opposite to $\vec{k}$, turns out to be described by the following dressed dielectric constant [15,16]

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

where 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 ($\beta \simeq 10^{-8}$ eV^2 at a pump power of 10 kW/cm^2). All the CuCl material parameters appearing in Eq. (1), i.e., the exciton and biexciton $\vec{k} = \mathbf{0}$ 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 experiments and are reported in Table I. The exciton and biexciton linewidths have been introduced phenomenologically and the contribution of the Z_{12} exciton resonance at 3.267 eV [18] included into the background dielectric constant ε_b. Small pump-induced nonresonant energy shifts [11] have been neglected as unimportant here.

The expression for ε in Eq. (1) has the typical three-level EIT form [1,19], the main features of which are also confirmed by recent calculations [5] going 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 ($\beta \rightarrow 0$), one obtains the usual upper and lower polariton branches and a polaritonic stop band within which the probe is nearly completely reflected [9]. In typical atomic systems or in the Cu_2O case, instead, the peak exciton absorption given by $\varepsilon_b \Delta_{LT}/\gamma_x$ is about 4 orders of magnitude smaller than in the present case and, thus, the width of the polaritonic stop band is negligible. The presence of the exciton-biexciton coupling through the pump introduces a third dispersion branch in the frequency region of the exciton resonance and this is expected to affect the probe reflectivity [11,15,20]. Our predictions show that transmission within the forbidden stop band can indeed be completely controlled.

Because several polariton branches are present, the knowledge of the local dielectric function ε is not sufficient to calculate the probe transmission through a

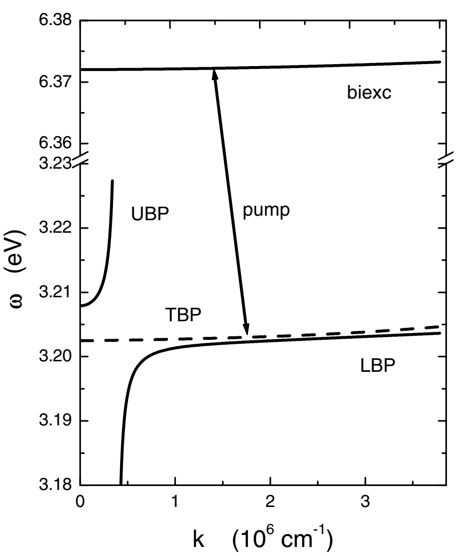

FIG. 1. Probe dispersion branches of coherently dressed CuCl. In the presence of the exciton-biexciton coupling pump, a third branch appears (dashed line) besides the usual upper and lower polariton ones.

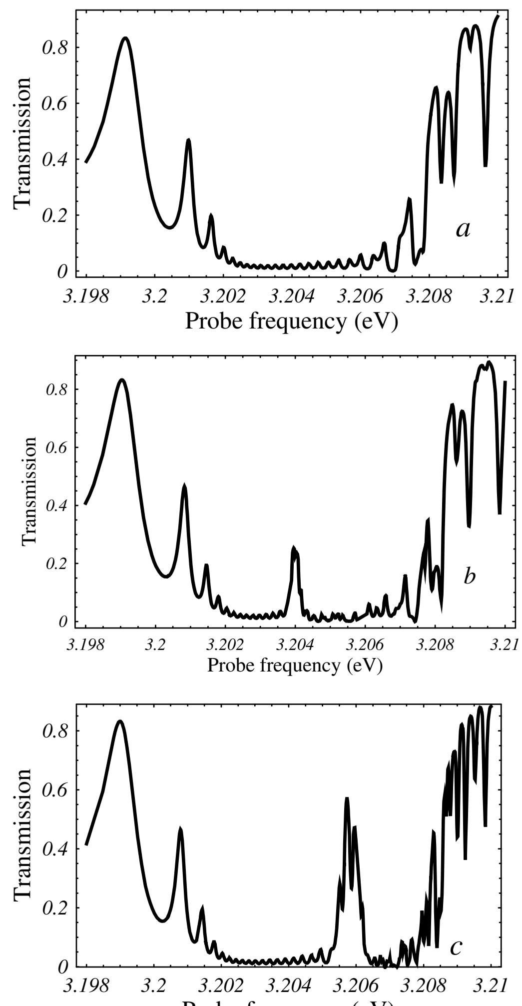

FIG. 2. Probe transmission spectra through a CuCl film 0.15 μm thick: (a) no pump, (b) with a pump of frequency $\hbar\omega_c = 3.168$ meV and intensity corresponding to $\beta = 5 \times 10^{-7}$ eV^2, (c) with a pump of frequency $\hbar\omega_c = 3.166$ meV and $\beta = 10^{-6}$ eV^2. The exciton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

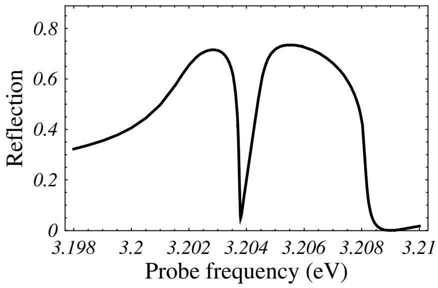

FIG. 3. Probe reflection for the case of a very thick sample ($d = 15\ \mu$m) with a pump of frequency $\hbar\omega_c = 3.168$ eV and $\beta = 10^{-6}$ eV^2. Other parameters are as in Fig. 2.

coherently driven slab of CuCl and the solution of Maxwell's equations requires additional boundary conditions (ABC) [9,11]. Here, we adopt the simplest ones, i.e., the extended Pekar's ABC [16] 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 [16].

The relevant probe dispersion branches [21], obtained from solving the equation $(c^2k^2/\omega^2) = \varepsilon(k, \omega)$ for a pump of frequency $\omega_c = 3.1695$ eV and intensity corresponding to $\beta = 10^{-7}$ eV^2, are shown in Fig. 1 for the case in which all dampings are neglected. The pump-induced new branch and the lower polariton branch anticross. We show instead in Fig. 2 the probe 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 $\omega \simeq \omega_m(k - k_c) - \omega_c$, similarly to the usual EIT two-photon resonance condition. Unlike in the case of EIT in atoms, 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 significant. In slabs thicker than a few microns, already representing the bulk limit (opaque slab), the effect of the exciton-biexciton coupling beam is best seen in reflection, as shown in Fig. 3.

Unlike the exciton linewidth, the biexciton linewidth at a small wave vector γ_m is seen to affect critically the appearance of a transparency window, as expected in analogy with EIT in the ladder configuration in atomic systems, and this is clearly shown in Fig. 4. Conversely, EIT in CuCl may also turn out to be a tool for the study of the polarization dephasings in the optical transition between exciton and biexciton states. Such a dephasing at low temperatures is largely governed by the radiative decay of the biexciton molecule and to a smaller extent by incoherent phonon scattering. Because the induced stop-band transparency in CuCl is quite sensitive to the biexciton dephasing, transmission measurements under EIT conditions can in turn precisely monitor the exciton molecule dephasing commonly measured by means of standard techniques based on the photoluminescence dynamics [13,22].

Finally, we have considered in Fig. 5 the delay experienced by a narrow probe Gaussian pulse propagating in the stop-band region of a coherently dressed thin slab. The pulse is 20% transmitted across the stop band, while from its delay one can infer a group velocity $v_g \simeq 5.8 \times 10^{-5}c$. This turns out to be in good agreement with the slope of the third dispersion branch induced by the exciton-biexciton coupling and which most contributes to the pulse propagating within the sample. Because the temporal pulse duration within the medium remains essentially unchanged, the pulse spatial length in the medium L_{in} is expected to scale approximately with the ratio v_g/c with respect to its length L_{out} in vacuum, and the remarkable spatial compression typically experienced by a pulse entering a slow-light medium [2] may also be observed in CuCl. As a consequence of such a spatial compression, an enhanced ponderomotive effect on free charges (electrons or holes) is expected to take place as the longitudinal gradient force exerted by the pulse scales with the inverse of the pulse length [23].

In conclusion, we have studied polaritonic effects associated with electromagnetically induced transparency

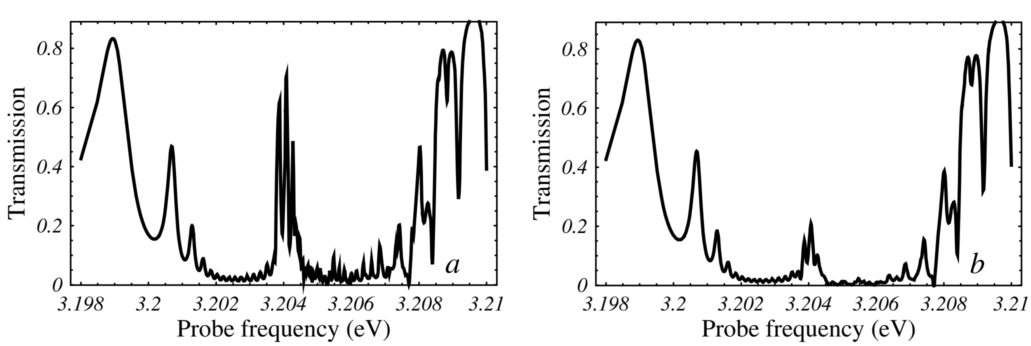

FIG. 4. Probe transmission spectra for different biexciton linewidths (a) vanishing γ_m, (b) $\gamma_m = 30\ \mu$eV. Pump frequency $\hbar\omega_c = 3.168$ eV and $\beta = 10^{-6}$ eV^2, while other parameters are as in Fig. 2.

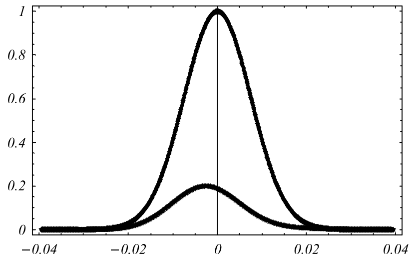

FIG. 5. Pulse profile in vacuum and after transmission within the transparency window induced by a pump of frequency $\hbar\omega_c = 3.168$ eV and $\beta = 10^{-6}$ eV^2 (other parameters as in Fig. 2). The peak lag of 2.6 mm corresponds to $v_g/c \simeq 5.8 \times 10^{-5}$.

in the realistic case of a thin slab of CuCl, and shown how a coupling beam resonant with the exciton-biexciton transition induces a transparency window for a probe beam within the polaritonic stop band. The probe transmission which is very sensitive to the biexciton dephasing can be coherently controlled changing the pump beam intensity and frequency over a wide range of values. A narrow pulse propagating within the transparency window suffers a spatial compression and a temporal delay corresponding to a reduction of its group velocity by 4 orders of magnitude.

Financial support from MIUR (Grant No. PRIN 2002-28858) is gratefully acknowledged.

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