Measurement of the Damping Dispersion of Exciton Polaritons in CdS

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At 2 K, at low intensity, and in good-quality samples, the source of the exciton-polariton dipole dephasing time (or phase-coherence time) is found to be extrinsic in nature and due to dephasing elastic collisions for excitation below the transverse-A-exciton frequency, and purely intrinsic and due to quasielastic collisions with acoustical phonons for excitation above the longitudinal-A-exciton frequency. The damping increases linearly with detuning for excitation above the longitudinal-A-exciton frequency.

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In this Letter, we report detailed measurements and a comprehensive study of the damping dispersion of both the A and B exciton polaritons in CdS over a wide frequency range. We identify the source of the damping and we demonstrate for the first time that, at low temperatures and in high-quality samples, the source of the damping is completely different for excitation above and below the lowest transverse-exciton frequency (ω_{TA}). It is found that the damping below the ω_{TA} frequency is extrinsic in nature and is governed by the impurity (and point defects) content of the sample. Above the ω_{TA} resonance, the dephasing is intrinsic and the results can be well explained by an exciton–acoustic-phonon interaction through the deformation-potential mechanism. This leads to a damping coefficient above the ω_{TA} frequency proportional to the detuning from the ω_{TA} resonance. These results challenge the common use of a constant, detuning-independent, phenomenological damping coefficient in the expression of the exciton contribution to the complex dielectric function. Our approach for extracting the exciton-polariton damping is based on the precise measurement of the optical transmission of high-quality CdS platelets. By working in a spectral region away from the longitudinal (ω_L) and transverse (ω_T) excitons, the present study deliberately avoids the need of determining the correct additional boundary condition and the appropriate dead-layer thickness.[1,2] Given the sample thickness and purity used in the experiment, the additional wave is of too small amplitude to perturb significantly the observed Fabry-Perot fringes. Future work in and around the ω_{LT} region should provide a means of testing different additional boundary conditions, as was already suggested by Broser, Pantke, and Rosenzweig,[3] but this is not the present intent.

We have restricted our present studies in CdS to light polarized perpendicular to the c axis and to frequencies around the A and B excitons. The dispersion relation, expressed by the dielectric function $\epsilon(k,\omega)$, is given by the following expression[4-6]:

$$\epsilon(K,\omega)=c^2K^2/\omega^2(K)=(n+i\kappa)^2 \tag{1}$$

$$=\epsilon_b+\frac{4\pi\beta_A\omega_{mA}^2(K)}{\omega_{mA}^2(K)-\omega^2(K)-i\omega(K)\Gamma_A}+\frac{4\pi\beta_B\omega_{mB}^2(K)}{\omega_{mB}^2(K)-\omega^2(K)-i\omega(K)\Gamma_B}, \tag{2}$$

where $\omega_{mA}(K)$ and $\omega_{mB}(K)$ are given by

$$h\omega_{mA}(K)=E_{mA,T}+\hbar^2K^2/2M_{xA} \tag{3a}$$

$$h\omega_{mB}(K)=E_{mB,T}+\hbar^2K^2/2M_{xB}\pm\phi K; \tag{3b}$$

κ is related to the absorption coefficient α by

$$2\omega\kappa/c=\alpha, \tag{4}$$

and n is the index of refraction. $E_{mA,T}$ and $E_{mB,T}$ are the mth transverse-exciton energies, β_A and β_B are the A and B exciton polarizabilities, ϵ_b is the background dielectric constant, which contains contributions from all interactions except the A and B excitons, and Γ_A and Γ_B are the damping coefficients of the A and B polaritons, respectively.

Over the years, it has been customary to use a phenomenological constant damping coefficient in the expression for the dielectric function.[5-7] This has been satisfactory since very few experiments require a detailed knowledge of the functional dependence of the damping. The techniques that have been used to study polaritons include reflectance, transmittance, resonant Brillouin scattering, photoluminescence, and nonlinear techniques such as four-wave mixing and hyper-Raman scattering. To our knowledge, the first convincing experimental evidence for a detuning dependence of the damping came from the work of Yu and Shen in Cu_2O.[8] In their experiment, it was found necessary to introduce a wave-vector-dependent damping term for the $1s$ yellow exciton to explain the dispersion of their Raman cross section. More recently, in the work by Ulbrich and Weisbuch in GaAs,[9] and Shigenari, Lu, and Cummins in CdS,[10] a significant broadening of the Brillouin peaks

was observed as the laser was scanned from below to above the transverse-exciton frequency ω_T. From such measurements, Shigenari, Lu, and Cummins extracted a complicated detuning dependence of the exciton-polariton damping. At frequencies more than 20 cm^{-1} above the ω_T frequency, a rapid decrease in the resonant Brillouin-scattering efficiency as well as increasingly serious local heating by their incident beam ($\simeq$8mW) made reliable measurements of the linewidth very difficult. For excitation below the ω_T or above the ω_L frequencies, the source of the damping was not clearly identified. Very recently, Broser, Pantke, and Rosenzweig monitored both the transmission and the reflection of a CdS sample and extracted the detuning dependence of the damping around the A-exciton resonance.[11] They claimed that the damping coefficient just around the resonance has the same detuning dependence as the group velocity of the A polariton. They speculated that the frequency dependence of the damping can be explained by the collision of polaritons with charged impurities. The role of acoustical phonons was completely ignored. In a different approach, Masumoto, Shionoya, and Takagahara[12] reported a nonlinear technique based on four-wave mixing for extracting the frequency dependence of the dephasing time of exciton polaritons in CuCl. Unfortunately, their technique failed to measure the low-intensity (concentration independent) dephasing time of exciton polaritons.

In our experiment, a krypton-ion–pumped cw ring dye laser operating with coumarin 102 dye around 487 nm is used as the source. The laser has a multimode linewidth of about 0.6 cm^{-1}. A photodiode monitors a small fraction of the 0.25-mW incident input power and another photodiode placed beyond the sample monitors the transmitted beam. The laser is scanned continuously with a computer-controlled stepping motor that turns the birefringent filter inside the dye laser. The wavelength of the light is accurately monitored by a Burleigh wavemeter and this number is stored in memory at the same time as the transmission is read and stored in memory. The CdS samples are held stress-free in superfluid helium. A recording of the transmission of a good optical-quality 1.2-μm-thick sample around the A and B excitons is shown in Fig. 1. The Fabry-Perot transmission oscillations are due to Fresnel reflection from the surfaces of the sample. A numerical fit that uses the theory to be described below is shown. Very good agreement between theory and experiment is obtained. The value of the physical parameters extracted from the best fit to the transmission data are shown in Table I.[13] e_{LT} is the longitudinal-transverse exciton splitting and f is the oscillator strength of the free exciton per unit cell of the crystal. The standard expressions for a Fabry-Perot etalon with losses were used.[14] There is a substantial literature of optical studies of CdS designed to extract the parameters of exciton polaritons.[15] These studies are in good agreement with the present results after due account is taken of Ref. 13. Even though many parameters are involved in the fit, they each have different effects on the spectrum.[1] The small uncertainty on the parameters shown in Table I is a measure of the accuracy of our fitting procedure when applied to different samples.

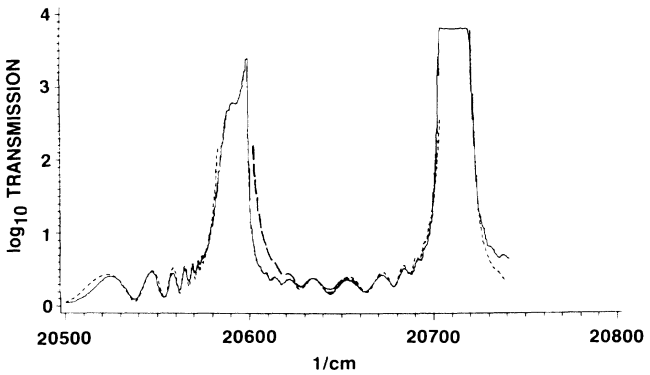

FIG. 1. Measurement of $\log_{10}$(transmission) vs $1/\lambda$ (solid line) for a high-quality 1.2-μm-thick CdS sample for frequencies around the A and B excitons. The short-dashed line is a theoretical fit to the experimental data with use of a detuning-dependent dephasing time above the ω_{TA} region. Good agreement is obtained. The long-dashed line is a fit under the assumption of a constant dephasing time (T_2=5 ps) above the ω_{TA} region. As can be seen, a single-value dephasing time only fits the experimental results over a very small energy range.

For excitation above the ω_{LA} frequency, the inner-branch polaritons are mostly responsible for the absorption process (conversion of the incident photon to a polariton). Subsequent to the absorption process, the inner-branch polariton is scattered to an outer-branch polariton by an acoustic phonon via the deformation-potential interaction. The total number of collisions per unit time W of an upper-branch polariton with a phonon system at 0 K is given by[16]

$$W(K)=(C_v-C_c)^2K^3/2\pi\rho c_s\hbar v_{Gl}, \quad (5)$$

where c_s is the LA phonon velocity, v_{GL} is the group velocity of the lower branch polariton, C_v and C_c are the deformation potential of the valence and conduction

TABLE I. Physical properties of CdS measured in our experiment.

$f_{A\,\mathrm{exc}}=2.30\times10^{-3}$
$f_{B\,\mathrm{exc}}=1.68\times10^{-3}$
$E_{TA}=20\,583.8\pm0.5\ cm^{-1}$
$E_{TB}=20\,706.0\pm1.0\ cm^{-1}$
$n_b=\epsilon_b^{1/2}=2.40\pm0.1$
$E_{LT}\vert_{A\,\mathrm{exc}}=15.8\pm0.3\ cm^{-1}$
$E_{LT}\vert_{B\,\mathrm{exc}}=14.5\pm0.3\ cm^{-1}$

TABLE II. Measurement of the A and B exciton-polariton dephasing rates in different samples for different laser detunings.

Sample thickness (μm)	Dephasing time below A exciton (ps)	Dephasing speed of the A exciton above the A exciton resonance (cm/s)	Dephasing speed of the B exciton below the B exciton resonance (cm/s)
12.5	100	1.9×10^9	9×10^8
2.8	28	2.1×10^9	8×10^8
15.6	100	1.8×10^9	7×10^8
1.21	40	3.8×10^9	1×10^9

bands (for CdS, C_v and C_c are tensorial entities), ρ is the crystal density, and K is the polariton wave vector in its final state. The exciton-polariton damping is directly related to the phonon collision rate by $\Gamma(K)=2W(K)$ or to the exciton-polariton dipole dephasing time by $\Gamma(K)=2/T_2(K)$. For large values of K ($K\gg M_xC_s/\hbar$), the damping is directly proportional to the detuning from the ω_T frequency. Even though CdS is an anisotropic crystal, we can approximately evaluate the importance of the LA phonon-scattering mechanism by using the following parameters: $C_v-C_c\simeq3.0$ eV, $M_x=1.3m_e$ (based on the average mass of an exciton in an ellipsoidal band), $\rho=4.82$ g/cc, $c_s=4.2\times10^5$ cm/s. For large K, we can define a dephasing speed v_ϕ in the following way:

$$W(K)=v_\phi\Delta\sigma, \tag{6}$$

where $\Delta\sigma$ is the detuning in reciprocal centimeters. We calculate a dephasing speed of $v_{\phi\text{theor}}=7.0\times10^8$ cm/s compared to the experimentally measured dephasing speed of $v_{\phi\text{expt}}=1.9\times10^9$ cm/s. The agreement is quite reasonable considering the uncertainty in the values of the deformation-potential parameters in CdS.[17]

For excitation below the ω_{TA} frequency, we have recently worked out a theory to explain the dephasing of exciton polaritons from elastic collisions with impurities and point defects. We have found that polaritons dephase at a rate[18]

$$W=\frac{1}{T_2}=N_{\text{imp}}\theta v_G\left[1-\frac{\sin(2\pi\Delta l/v_GT)}{2\pi\Delta l/v_GT}\right], \tag{7}$$

where v_G is the polariton group velocity, n_{imp} is the impurity concentration, θ is the impact cross section, and Δl is an effective length that characterizes the collision. For small v_G, W is proportional to v_G. For large v_G, the dephasing rate W is inversely proportional to v_G. When different types of impurities contribute to the polariton scattering, a phenomenological, sample-dependent, constant dephasing rate often provides a reasonable fit to the transmission data in the spectral region of interest. Table II shows the values of the parameters used to characterize the dephasing both above and below the transverse-exciton frequency in four different samples. As can be seen, widely different dephasing times are required to characterize the absorption below the frequency ω_T in different samples. On the other hand, above ω_T the dephasing speeds are found to vary little from sample to sample, indicating the intrinsic nature of the process responsible for dephasing the polaritons. Only in the thinner sample, where thermal effects are more likely to play a role, do we observe a larger dephasing speed. Indeed, a temperature of only 5 K is sufficient to explain this result. A temperature study revealed that the damping below ω_T is independent of temperature up to 50 K and that the damping above ω_T is a very sensitive function of temperature. These temperature results also support the extrinsic nature of the damping below ω_T and the intrinsic nature of the damping above the transverse-A-exciton frequency.

In conclusion, detailed transmission measurements have clearly identified the extrinsic origin of the damping below the A free exciton and the intrinsic role of acoustical phonons in the dephasing of polaritons above the transverse-exciton frequency. Precise values of the dephasing rate of exciton polaritons were obtained over a very wide frequency range around the A and B excitons in CdS.

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