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Polariton Wave Packet Propagation in the Exciton Resonance of a Semiconductor

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This paper reports the first experimental observation of "slow" pulse propagation in the exciton-polariton resonance of a direct-gap semiconductor. The propagation delay of short light pulses transmitted through a 3.7-μm-thick GaAs crystal was measured directly with a cross-correlation technique. Polariton group velocities from $c_{vac}/3.6$ down to $c_{vac}/2000$ were found when the wave-packet center frequency was tuned through the $n=1$ exciton resonance.

As a consequence of the Coulomb interaction the low-lying electronic excitations in semiconductors are modified: The electron-hole correlation causes discrete exciton resonances and electron-hole–pair continuum states. Their coupling to the light field (which is generally strong in the case of direct allowed optical transitions), is the essential in the concept of the polariton, a mixed exciton-photon state, which corresponds to dielectric polarization in the macroscopic description of dielectrics.[1] The classical problem of wave propagation *in* and transmission *through* such bounded "spatially dispersive" dielectric media for frequencies around the exciton resonances has recently attracted renewed interest in the context of transient phenomena.[2,3] Up to now, the continuing theoretical work towards a consistent description of the problem (including the interpretation of resonant light scattering spectra) in terms of proper dielectric functions and physically correct boundary conditions[4,5] has not been adequately consolidated by selective experiments on well-defined crystals.

We present in this Letter the result of a conceptually simple, but nevertheless fundamental, experiment on polaritons: the first direct study of the enormous variation of the group velocity around a discrete exciton resonance done by measuring the propagation delay of wave packets transmitted through a thin plane-parallel slab.

The GaAs samples of controlled thickness $d=3.7$ μm and $\sim 200\times 500$ μm^2 size were prepared by standard lapping and etching from high-purity ($N_D, N_A \leq 5\times 10^{14}$ cm^{-3}) vapor-phase epitaxy material. Figure 1(a) shows the experimental arrangement: Bandwidth-limited light pulses of 12-psec duration and 0.2-meV spectral width ($\Delta\nu \cdot \Delta\tau = 0.6$) with accurately adjustable center frequency ω_0 were generated with a synchronously mode-locked oxazine-750 dye laser.[6] The laser beam was split into reference and probe beams; the latter was focused into a 50-μm-diam spot giving ~ 1 kW/cm^2 pulse peak power incident on the sample, which was held in pumped liquid He at 1.3 K. The chosen pump power level was checked carefully to be well below the onset of nonlinear bleach-

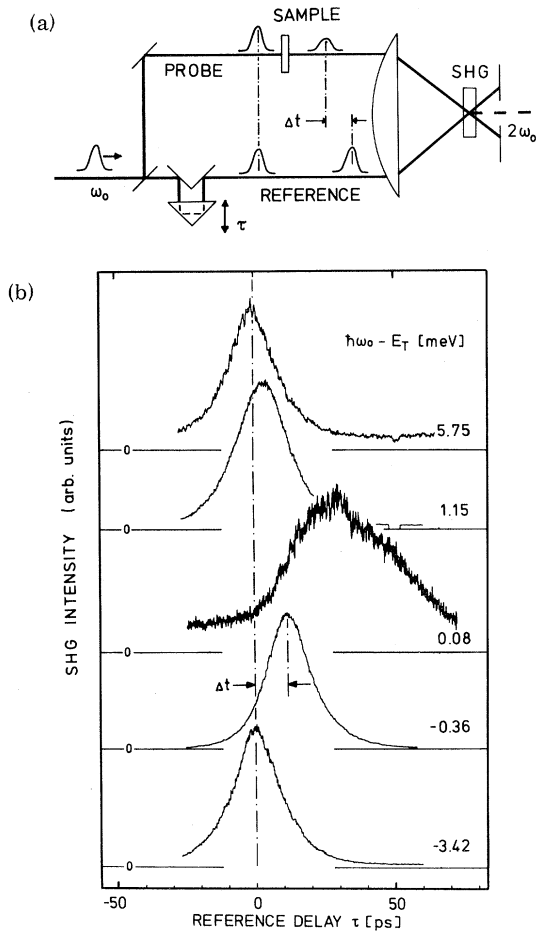

FIG. 1. (a) Experimental arrangement for measuring the propagation delay of picosecond light pulses transmitted through a thin sample. Noncollinear second-harmonic generation (SHG) in $LiIO_3$ is used to perform the envelope-intensity cross correlation between probe and reference pulses. (b) The cross-correlation (SHG) signal for five different light-pulse center frequencies ω_0 around the exciton resonance at E_T in the direct-gap semiconductor GaAs at $T = 1.3$ K. The light pulses were bandwidth limited and had a 12-psec duration.

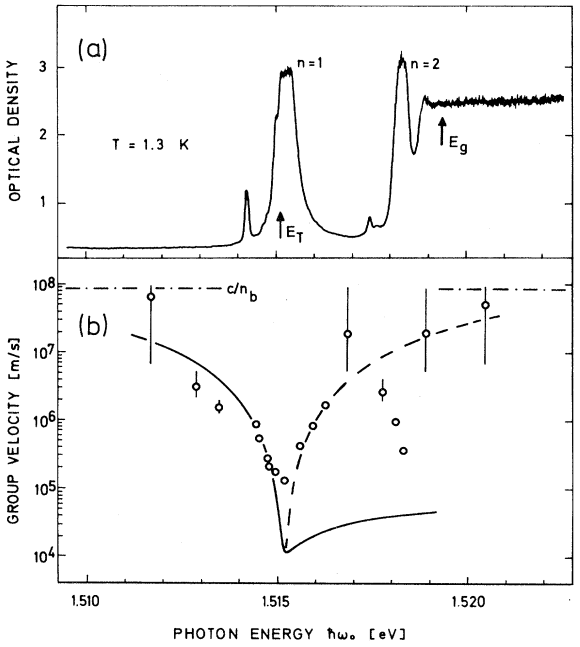

FIG. 2. Optical density spectrum of the 3.7-μm-thick GaAs sample in the region of discrete exciton resonances $n = 1, 2...$, obtained from a standard cw white-light absorption experiment. E_T denotes the lowest transverse exciton energy, E_g the band-gap energy (onset of continuum). (b) Experimental polariton wave-packet group velocities on the same energy abscissa scale (note the logarithmic ordinate scale!). The lines are the theoretical group velocities $\partial\omega/\partial k$ for the two polariton branches of the two-band single-exciton oscillator model according to Ref. 1, taking the known parameters of Ref. 10 (solid line, lower branch; dashed line, upper branch). c/n_b denotes the phase velocity caused by the background dielectric constant.

ing and saturation phenomena in the exciton absorption line.[7]

The propagation delay of the light pulses transmitted through the sample was measured by means of a cross-correlation technique utilizing type-I noncollinear second-harmonic generation (SHG) in $LiIO_3$ (Ref. 8) with the adjustable time delay τ in the reference beam and detection of the SHG signal by a photon-counting system. The geometry and polarization vectors were chosen such that the time-averaged SHG signal at $2\omega_0$ is given by

$$\bar{\mathcal{F}}_{\mathrm{SHG}}(\tau) \propto \lim_{T\to\infty} T^{-1}\int_0^T |\vec{E}_{\mathrm{probe}}(t)|^2\,|\vec{E}_{\mathrm{ref}}(t+\tau)|^2\,dt\,, \tag{1}$$

i.e., the envelope of the transmitted probe pulse intensity folded with the unperturbed reference pulse. The accuracy of the envelope delay measurements was restricted to about 0.2 psec (0.5 psec in the region of higher damping) by laser intensity fluctuations and shot noise at low transmitted intensities.

To obtain independent information on the spectral damping of the GaAs samples,[9] a cw absorption spectrum was measured with well-collimated broad-band (tungsten-lamp) excitation and 0.1-meV resolution under otherwise identical condi-

tions [see Fig. 2(a)]. The $n=1$ and $n=2$ exciton resonances are well resolved; the structure at 1.5142 eV is the (D^0,X) donor-bound exciton. The spectrum shown has a noise-limited dynamic range of $0 \lesssim p_{\mathrm{opt}} \lesssim 3$ due to low intensity excitation and is not corrected for the ω-dependent reflectivity at the sample surfaces. No significant changes in the optical density were observed as compared to the picosecond excitation. The energetic position of the transverse exciton energy, $E_T = 1.5151$ eV,[10] is indicated in Fig. 2(a).

Tuning the light-pulse center frequency ω_0 into the exciton resonance at E_T, a monotonic increase in pulse propagation delay (reaching ~35 psec at the maximum) was observed. Figure 1(b) shows five of the individual pulse-envelope cross-correlation traces, which actually were taken in closely spaced intervals around E_T and showed no significant change in pulse *shape* unless $|\hbar\omega_0 - E_T| < 0.3$ meV. Within this interval the mean delay was quite large and the correlation signal became asymmetric with a trailing edge "tail."

From the correlation data [Fig. 1(b)] we determined the relative time delay Δt of the first moment of the pulse intensity distribution:

$$\Delta t = \int_{-\infty}^{+\infty} \tau \mathfrak{F}_{\mathrm{SHG}}(\tau)\, d\tau \Big[\int_{-\infty}^{+\infty} \mathfrak{F}_{\mathrm{SHG}}(\tau)\, d\tau \Big]^{-1}. \qquad (2)$$

The circles in Fig. 2(b) show the measured propagation velocity v_{gr} of the wave packet through the sample, defined by

$$v_{\mathrm{gr}} = d/\Delta t\,. \qquad (3)$$

The spectral bandwidth of the pulses is given by the circle diameter and the vertical error bars indicate the envelope timing uncertainty equivalent to 0.5 psec in Δt. Possible multiple-reflection effects in the plane-parallel sample, which lead to pulse delay and stretching, can be neglected here because of the relatively high damping in the spectral region of interest [see Fig. 2(a)].

The lines in Fig. 2(b) show the predicted dependence of the polariton group velcoity $v_{\mathrm{gr}} = \partial\omega/\partial k$ (full line, lower branch; dashed line, upper branch) in the two-band single-exciton oscillator model[1] with use of the values $E_T = 1.5151$ eV, $E_{\mathrm{LT}} = 0.08$ meV (longitudinal-transverse splitting), $M_{\mathrm{EX}}{}^h = 0.6 m_0$ (heavy-exciton mass), and $n_b = 3.55$ (background dielectric constant) known from resonant Brillouin scattering experiments.[10]

The agreement with the experimental points is good and displays strikingly the enormous change in group velocity of about three orders of magnitude when ω_0 is tuned through the resonance. The data are an impressive manifestation of the renormalization of excitons at E_T due to their coupling to the photon field!

In the spectral region of spatial dispersion of the $n=1$ exciton, $|E_T{}^{1s} + E_{\mathrm{LT}}| < \omega_0 < E_T{}^{2s}$, we clearly observe pulse propagation with $\partial\omega/\partial k$ corresponding to the *upper* (i.e., photonlike) *polariton branch*. This fact directly tells that the contribution of the lower (i.e., excitonlike) branch to energy transport is negligible either because its damping is so large or because its initial (and final) coupling to the external light field at the crystal boundaries is so small. The same argument applies for the question of relevance of the third polariton branch originating from the *light* $\Gamma_6 \otimes \Gamma_8$ exciton band.[11] The data points of Fig. 2(b) also show clearly the $n=2$ exciton resonance (which is not considered in the theoretical curves).

The pronounced asymmetry in the cross-correlation spectrum for $|h\omega_0 - E_T| < 0.3$ meV [see Fig. 1(b)] is very probably due to the strong variation of $\partial\omega/\partial k$ *and* damping in this narrow spectral region. The chosen bandwidth of 0.2 meV is no longer small on this scale and leads to strong pulse distortion by selectively damping and dephasing parts of the pulse frequency spectrum.

A careful inspection of the cross-correlation pulse shape *wings* revealed small distortions and asymmetries in the trailing and leading edges [not visible in the linear plots of Fig. 1(b)]. We could not yet ascribe unambiguously these structures to distinct propagation modes, which were discussed recently.[2] Unless the full incident pulse spectrum (in time *and* frequency domain) is stable and known with high accuracy, the search for separate wave fronts (the "precursors" of Ref. 2 or the beating phenomena of Ref. 3) with small amplitudes is difficult, though in principle feasible with the state-of-the-art picosecond techniques applied in our experiment.

In conclusion, we have reported the result of a conceptually simple experiment which manifests impressively the effect of renormalization of excitons through their strong coupling to light: enormous group-velocity variation (over three orders of magnitude) controlling the polariton wave-packet propagation around the lowest $n=1$ exciton resonance in the model semiconductor GaAs. Our measurements of picosecond light-pulse delays after transmission through thin crystals verified for the first time this effect, which has been anticipated long before this experiment could actually be performed.[1]

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Crossover from Negative to Positive Spin Polarization in the Photoyield from Ni(111) near Threshold

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The measured spin polarization of the photoelectrons emitted from Ni(111) shows a crossover from negative to positive values at 270 ± 90 meV above photothreshold. This is in constrast to the case of Ni(100), where a crossover at about 75 meV occurs. A single-particle photoemission calculation with an exchange splitting of 0.33 eV, and which does not invoke emission from surface states, is in good agreement with the experimental results for both faces. These new results corroborate the empirical band structure obtained by Eastman and co-workers from angle-resolved photoemission data.

In this Letter, we report measurements of the electron spin polarization (ESP) of photoemitted electrons from a well-characterized single-crystal Ni(111) surface for photon energies $\hbar\omega$ ranging from threshold φ to about 1 eV above φ. We find that the spin polarization changes from negative values (− 45% at threshold, i.e., preferential direction of the magnetic moment of the emitted electrons antiparallel to the magnetization) to positive values at an energy $E_c = 270 \pm 90$ meV above threshold. A photoemission calculation based on the Stoner-Wohlfarth-Slater (SWS) theory and using an exchange splitting of 0.33 eV is in good agreement with the experiment. This allows us to extract the value of the exchange splitting and the value of the Stoner gap for the ⟨111⟩ direction. In agreement with the interpretation of recent angle-resolved ultraviolet photoemission spectra[1] (ARUPS), surface-state emission is found to be not necessary to interprete the experimental ESP data for both (100)[2] and (111) surfaces studied to date in contrast to the suggestion by Dempsey and co-workers.[3]

Ni is accepted to be the prototype "strong" ferromagnet i.e., the majority-spin d band is full and the Fermi level crosses only the minority-spin d band. Primarily two related quantities characterize the electronic structure of Ni: the magnetic exchange splitting Δ and the "Stoner gap" δ, the latter being the energy differences