Observation of optical precursors at pulse propagation in GaAs

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We have observed optical precursors by studying propagation of ultrashort optical pulses with a steeply rising and an exponentially decaying trailing edge through a 0.2-μm-thick GaAs crystal near the exciton resonance. The appearance of a fast pulse front—the precursor—independent of the detuning with respect to the resonance, was accompanied by a delayed main pulse. The delay of the main pulse varies with detuning in accordance with theoretical predictions.

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The appearance of precursors in the transient response of linear dielectric media to the ultrashort pulse excitation was first theoretically predicted in 1914 by Sommerfeld [1] and Brillouin [1] who studied the propagation of a truncated harmonic wave $E(t,z=0)=\Theta(t)\cos\omega_L t$ in a dispersive dielectric, where $\Theta(t)$ is the Heaviside step function. It was shown that at a distance $z>0$ inside the semi-infinite medium a fast front of the pulse arrives at $t_s=(z/c)(\epsilon_\infty)^{1/2}$ (corresponding to the high-frequency components of the initial pulse) followed by another maximum at $t_B=(z/c)(\epsilon_\infty+4\pi\alpha)^{1/2}$ (due to the low-frequency part of the initial pulse) before the main pulse arrives at $t_g=z/v_g$. Here $v_g=\partial\omega/\partial k$ denotes the group velocity at the laser frequency ω_L and the dielectric function $\epsilon(\omega)$ is modeled by Lorentz oscillators according to

$$\epsilon=\epsilon_\infty+\frac{4\pi\alpha\omega_0^2}{\omega_0^2-\omega^2-i\omega\Gamma}\,, \qquad (1)$$

with ϵ_∞ the dielectric constant due to higher-lying resonances, ω_0 the frequency, Γ the width, and α the polarizability of the resonant transition.

The classical theoretical treatment of this transient electromagnetic wave propagation in local media is summarized in [2]. Later the precursor theory of Sommerfeld and Brillouin has been extended to spatially dispersive (nonlocal) media by Birman and Frankel [3]. In their work the existence of an additional slowly propagating exciton precursor, associated with the excitonic part of the lower polariton branch, has been suggested. Both the Sommerfeld and the Brillouin precursor have been detected experimentally for the propagation of microwave pulses in dispersive waveguides [4], whereas observation of optical precursors has not been reported so far. Experimental pulse propagation studies in the optical domain have been interpreted in the framework of the group-velocity concept [5], including pulse broadening and higher-order distortion effects. It is noteworthy that group velocities as low as $10^{-5}c$ have been observed [6].

In a recent paper [7] we analyzed the observability of optical precursors in the light of the present experimental facilities for generating ultrashort optical pulses. In that paper we proved by simultaneous temporal and spectral analysis of the transient response that detection of optical precursors preceding the main pulse can be accomplished by following the propagation of comparatively narrow-bandwidth pulse with a steep rising or falling edge and with a frequency tuned close to a relatively sharp material resonance.

In the present paper we report on the experimental observation of Sommerfeld and Brillouin precursors in the optical frequency range. Following the aforementioned considerations [7] we investigated the shape of an almost single-side exponential pulse with a frequency tuned close to the exction resonance after transmission through a thin GaAs layer. Independent of the detuning of the laser frequency with respect to the exciton resonance, we observed a sharp pulse front followed by the delayed main pulse. The delay and the shape of the main pulse as a function of the frequency detuning agrees quite well with the prediction of the simple theoretical model presented in [8].

The crucial point of the experiment is the single-sided exponential profile of the incident pulse corresponding to a Lorentzian line in the frequency domain, which is distinguished by broad wings. For such a pulse two limiting propagation regimes can be differentiated. First, the precursor part of the signal arises because of the nonresonant (high- and low-frequency) spectral components of the initial pulse, which are influenced only slightly by the resonance while traveling through the sample at the speed c/n_∞. Second, the group velocity can be ascribed to the spectrally narrow part of the pulse near the resonance, whose motion is slowed down by the resonance. In this case and for a weak material resonance (i.e., with a splitting between the longitudinal and transverse eigenfrequencies of the exciton $\Delta_{LT}=2\pi\alpha\omega_0/\Gamma<\Gamma/2$), the following characteristic time constants are important [7]: the rise time τ_R and the decay time τ_D of the initially incident pulse, the duration of the precursor spike T_S

$$T_S=\frac{(\epsilon_\infty)^{1/2}c}{4\pi\alpha\omega_0^2 z}\,, \qquad (2)$$

and the delay corresponding to the minimum of the group velocity

$$T_g = \frac{z}{v_g^{\min}} = \frac{4\pi\alpha\omega_0 z^2}{8(\epsilon_\infty)^{1/2} c\Gamma^2} . \tag{3}$$

Optimum conditions for observing the optical precursors, traveling well ahead of the main pulse, are realized if the characteristic times satisfy the relation

$$\tau_R \lesssim T_S \lesssim T_g \lesssim \tau_D , \tag{4}$$

and if

$$D = \frac{4\pi\alpha\omega_0^2 z}{4\epsilon_\infty c\Gamma} \cong 1 , \tag{5}$$

which requires that absorption in the sample is not too high at the frequency experiencing the maximum delay. Let us note that for the case of a weak resonance in the optical region $4\pi\alpha \ll \epsilon_\infty$, and, therefore, the Sommerfeld and Brillouin precursors are hardly distinguishable.

The sample under study was a molecular-beam-epitaxy (MBE) structure grown on an n^+ substrate and containing a 0.2-μm-thick GaAs layer sandwiched between two GaAs/$Al_{1-x}Ga_xAs$ superlattices. An additional 0.2-μm-thick $Al_{1-x}Ga_xAs$ layer was inserted between the lower superlattice and the n^+ substrate, providing sufficient selectivity for the etching process applied for removal of the substrate. The substrate was first polished down to 15 μm and next 0.5-mm holes were etched into the substrate leaving thin free layers of GaAs for transmission experiments. The transmission spectrum of the sample is given in Fig. 1. The $n=1$ exciton line at 12221 cm^{-1} has a full width at half maximum (FWHM) of 4 cm^{-1} of the corresponding absorption coefficient, the band-to-band absorption edge (together with higher exciton lines) is smeared and extends to the $n=1$ exciton line. The corresponding luminescence spectrum shows a resonant exciton emission line (FWHM of 4 cm^{-1}) and bound exciton lines due to neutral donors (at 12213 cm^{-1}) and neutral acceptors (at 12119 cm^{-1}). The free and bound exciton lines of the polished and etched samples are broader as compared to the linewidths in the as-grown samples which amount to 3

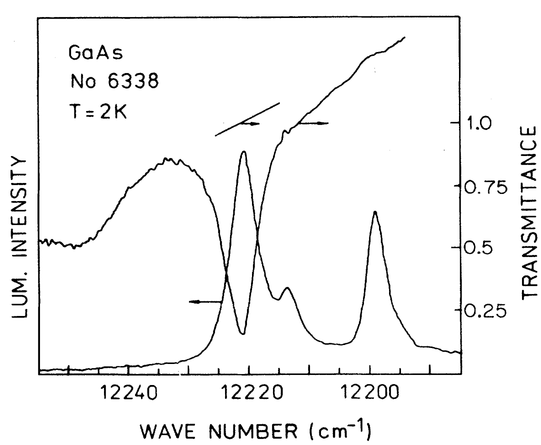

FIG. 1. The transmission (upper curve) and luminescence (lower curve) of the 0.2-μm-thick GaAs sample at $T=2$ K.

and 1 cm^{-1}, respectively. This indicates inhomogeneous broadening of the exciton levels due to strain induced by the sample preparation process. Experiments were carried out in liquid He pumped below the λ point.

The excitation source included a hybridly mode-locked Styryl-9M dye laser with hexamethylindotricarbocyanineiodide as the saturable absorber, pumped by the second harmonic of a neodymium-doped yttrium aluminum garnet (Nd:YAG) laser. This system delivered 200 mW of average power at 12220 cm^{-1}. Wavelength selection and tuning of the dye laser was accomplished by a single-plate birefringent filter. The autocorrelation trace had a FWHM of 700 fs and the corresponding emission spectrum was 40 cm^{-1} wide. Spurious spectral structures on a 1 cm^{-1} scale were observed due to unavoidable Fabry-Pérot resonances in the dye laser cavity. The dye laser output was split into two beams, one of them serving as the pump in the subsequent cross correlation measurement. The other beam was passed through a piezoelectrically tunable Fabry-Pérot étalon (97.5% reflectivity and 50 μm spacing of the mirrors). The incident pulse was thus convoluted with the almost single-sided exponential transfer function of the étalon corresponding to the Airy function transmission profile of the Fabry-Pérot étalon in the frequency domain. For our étalon we obtained a nearly exponential pulse with a rise time $\tau_R = 400$ fs and decay time $\tau_D = 6.2$ ps. The spectral width of the pulse behind the étalon amounted to 1.5 cm^{-1}. The 1-mW beam transmitted through the Fabry-Pérot étalon was focused onto the sample keeping the average incident power density below 1 W/cm^2. Time-resolved analysis of the pulse shape after transmission through the sample was attained via cross correlation [9] (up-conversion) with the initial laser pulse in a several millimeter long $LiIO_3$ crystal. The cross correlation function (CCF) of the transmitted pulse was analyzed by standard synchronous detection technique.

The CCF of the transmitted pulses measured for various values of detuning of the incident pulse with respect to the exciton resonance ω_0 are depicted in Fig. 2(a) together with the CCF of the incident exponential pulse (upper curve). The appearance of a fast, undelayed front of the transmitted pulse is clearly evident for all traces. It follows closely the front of the incident pulse and is observed independent of the detuning parameter $\Delta = \omega_0 - \omega_L$. This spike corresponds to the predicted precursor response and should be compared to the following maximum, which is considerably delayed with respect to the fast pulse front. The delay and the shape of the main pulse varies with the detuning Δ.

In Fig. 2(b) the theoretical CCF are given, which have been calculated by straightforward Fourier analysis using the dielectric function $\epsilon(\omega)$ with the following parameters: $\epsilon_\infty = 12.6$, $\omega_0 = 12220$ cm^{-1}, $4\pi\alpha = 1.3 \times 10^{-3}$, $\Gamma = 4$ cm^{-1}, and $z = 0.2$ μm for the medium, and $\tau_R = 500$ fs and $\tau_D = 5$ ps for the rise and decay times of the incident pulse, respectively. It is important to note that for a single Lorentzian resonance the transmitted pulse shape is independent on the sign of the detuning. The theory predicts a sharp rise of the transmitted response independent of the parameter Δ and a delay of the main pulse depend-

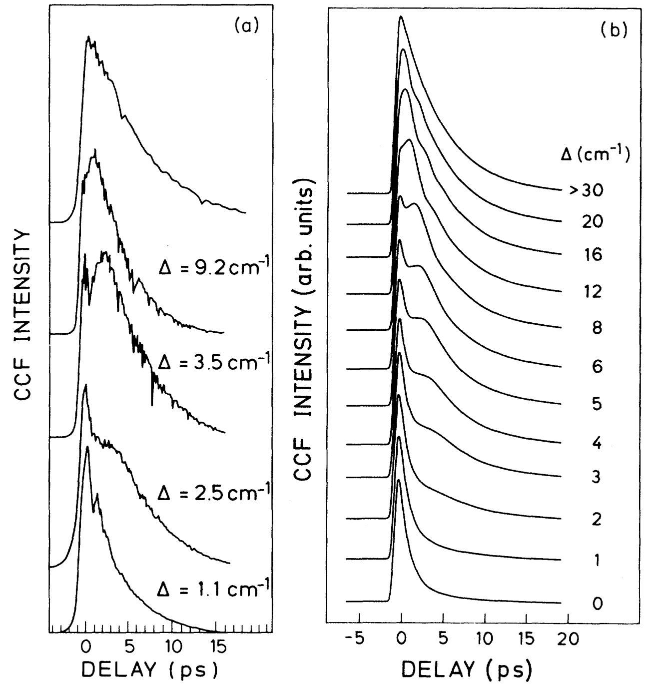

FIG. 2. The normalized CCF of the incident exponential pulse (uppermost curve) and transmitted pulses as a function of detuning Δ (in cm^{-1}); (a) experiment, and (b) theory (see text for parameters).

ing on Δ. At zero detuning a distinctly shortened transmitted pulse emerges. This pulse compression is caused by the anomalous dispersion of the refractive index at the resonance frequency. Comparing the experimental and theoretical CCF we see that the theory describes fairly well the qualitative features of the transmitted pulse revealing both the precursor and the delayed main pulse for $\Gamma/2 < \Delta < \Gamma$. The maximum of the main pulse shifts with detuning Δ and may be delayed up to several picoseconds with respect to the sharp front. For small detunings this shift cannot be directly related to the group delay T_g, which applies rather to the rising edge of the main pulse. This is especially clear for the anomalous dispersion region $|\Delta| < \Gamma/2$, where $dn/d\lambda < 0$ and the group-velocity index

$$n_g = n_\infty - \lambda dn/d\lambda$$

becomes smaller than n_∞ or even negative [10], and the transmitted pulse thus obtains a rather specific shortened shape. This feature is also experimentally observed, as well as the slightly oscillatory decay of the transmitted pulse at larger detunings. The good agreement between experimental results and theoretical predictions allows us to conclude that a coarse and rather straightforward model of an isolated resonance reasonably describes the pulse propagation near the excitonic resonance in GaAs. It is evident that the model cannot provide a quantitative description of the experiment since several effects which may influence the transient response are not included in the present treatment. First, spatial dispersion has been neglected. Second, the frequency dependence of the reflection coefficient on both sample surfaces, resulting in additional transients, is not considered. Third, the dead-layer problem associated with reflection near excitonic transitions in a semiconductor should be taken into account.

Nevertheless, we have tried to fit the experimental data numerically (see Fig. 3). A satisfactory fit could be obtained only if the product $4\pi\alpha z$ was increased by a factor of 1.5–2 with respect to the parameters used above. This misfit was also present when we measured directly the maximum group delay by analyzing the propagation of long (FWHM of about 8 ps) Gaussian pulses. The observed delay was about 700 fs, that exceeds 2.4 times the estimate $T_g = 290$ fs. This discrepancy may be related either to a miscalculation of z or to strain-induced inhomogeneities. Samples with a well defined and smaller homogeneous linewidth are required to elucidate this problem.

The experimental results represent a successful step towards the detection of precursors in the optical regime. Both the Sommerfeld and the Brillouin precursor contribute to the fast pulse front of the signal transmitted through the sample. The experimental approach does not permit separation of the high- and low-frequency precursors because $4\pi\alpha \ll \epsilon_\infty$ for the weak excitonic resonance. However, the present status of the technology for the generation of ultrashort optical pulses still constrains such studies to relatively weak and narrow resonances. The Fourier-transform-limited bandwidth of a 200-fs pulse amounts to approximately 50 cm^{-1}. Thus the influence of the resonance on the propagation of light through the crystal must be negligible for a detuning of a few 10 cm^{-1}. Investigations of the forerunner phenomenon near a strong electron resonance would require pulse rise times which are still several orders of magnitude beyond the present limit of a few femtoseconds (see our corresponding estimate in Ref. [7]).

Let us finally represent the CCF of the short (and spectrally broad) $sech^2$-shaped pulse transmitted through the

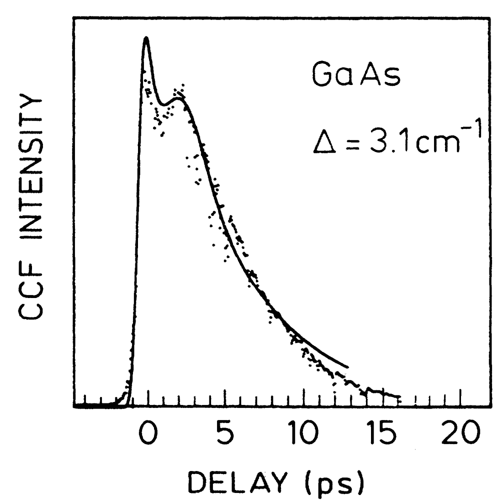

FIG. 3. The normalized CCF of the transmitted exponential pulse for detuning $\Delta = 3.1$ cm^{-1}. Solid line: theory ($4\pi\alpha = 2\times10^{-3}$); dots: experiment.

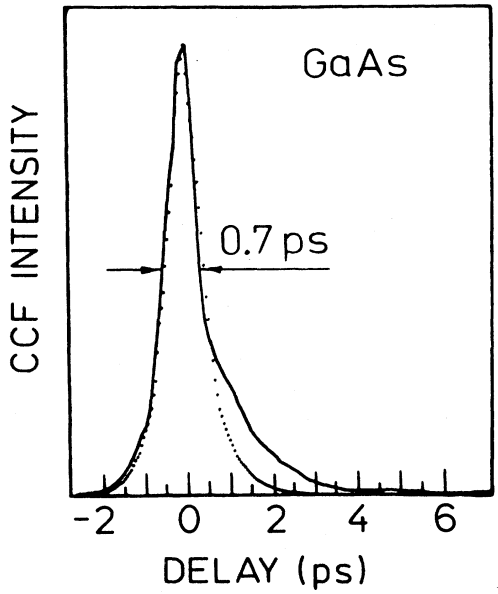

FIG. 4. The CCF of the short incident (dotted curve) and transmitted (solid curve) sech^2 pulses at $\Delta=0\ \mathrm{cm}^{-1}$.

sample at the resonance frequency (Fig. 4). A weak oscillatory tail is observed as a result of dispersion of the frequency components near the exciton resonance. This tail vanishes gradually when we detune the incident pulse. A similar experiment has been carried out in optically thick sodium vapor [11], where the formation of a long oscillatory 0π-area pulse was observed. In our case of an optically thin sample ($\alpha_0 l \approx 1$ in the terms of Ref. [11]) the reshaping of the incident pulse is essentially weaker and results merely in a weak tail.

In conclusion, we have experimentally demonstrated the existence of precursors for transient electromagnetic wave propagation through dispersive media in the optical frequency range. These precursors or forerunners appear in the transmitted optical pulse if long narrow-bandwidth pulses with steep fronts propagate near material resonances. Experiments on thin GaAs crystals with nearly exponential type pulses and a frequency tuned close to the free-exciton resonance have confirmed this prediction in a good agreement with corresponding theoretical calculations. The observation of separate Sommerfeld, Brillouin, and exciton precursors in the optical regime remains a challenge for future work.

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