Excitation of Coherent Phonon Polaritons with Femtosecond Optical Pulses

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Coherent lattice vibrations in lithium tantalate have been observed directly in the time domain by use of femtosecond optical pulses and the electro-optic effect to coherently excite and detect phonon polaritons. The damping time of the lowest transverse optic phonon has been determined directly from the decay rate of these oscillations. This measurement establishes an intrinsic speed limit for the electro-optic response of this material.

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Optical techniques have been widely used to study the lattice dynamics of solids. Linear spectroscopy in the far infrared and nonlinear optical and Raman spectroscopy have provided a wealth of information about the kinetics of phonons and their coupling to other elementary excitations in the condensed phase. These approaches have exploited the frequency response of the linear or nonlinear coupling of phonons to the electromagnetic field. Although extensive time-resolved measurements have been made with picosecond optical pulses, in most cases the detection is not phase coherent and measures only the envelope of the decay of the excitation. An exception is the coherent excitation and detection of acoustic phonons by picosecond impulsive stimulated Brillouin scattering.[1] Until recently, the temporal resolution of optical sources was not sufficient to resolve the individual oscillations of an optic phonon. With the development of femtosecond optical lasers,[2] it is now possible to have temporal resolution less than one cycle of an optic phonon. This has made possible coherent excitation and detection of optic phonons by impulsive stimulated Raman scattering.[3]

In this paper we report the first coherent impulse excitation and detection of phonon polaritons by the electro-optic effect in noncentrosymmetric crystals.[4] Our approach uses femtosecond optical pulses for both excitation and detection. Coupling to the TO phonon is accomplished by the electro-optic effect which has a strong resonant contribution due to ionic lattice displacements. The physical basis for our experiment can be illustrated by a coupled-oscillator model[5] which accounts for the interaction between the ionic and electronic displacements and the electromagnetic field. In this picture, the total polarization of the material, $P(t)$, is equal to the sum of ionic and electronic contributions, $P_i(t)$ and $P_e(t)$, each of which is determined by coupled oscillator equations:

$$\frac{\partial^2 P_i}{\partial t^2}+\Gamma_i\frac{\partial P_i}{\partial t}+\omega_i^2 P_i=\alpha_i E+\beta_i P_e^2, \quad (1)$$

$$\frac{\partial^2 P_e}{\partial t^2}+\Gamma_e\frac{\partial P_e}{\partial t}+\omega_e^2 P_e=\alpha_e E+2\beta_e P_e P_i, \quad (2)$$

where Γ_i and Γ_e are the ionic and electronic damping factors, ω_i and ω_e are the ionic and electronic resonant frequencies, α and β are linear and nonlinear coupling factors, and E is the total electric field. In each of the oscillator equations there are both linear and nonlinear driving terms. The ionic polarization has a nonlinear driving term proportional to the square of the incident optical polarization. This effect, known as optical rectification,[6] is used in our experiment to provide the impulse excitation of the lattice mode. The resulting polarization couples linearly to the electric field and radiates a low-frequency electromagnetic pulse. As previously demonstrated,[7] this produces a far-infrared pulse with a Cherenkov-type conical wave front. This pulse is strongly coupled to the lattice and propagates as a polariton, i.e., it is a mixed excitation having both photon and phonon properties. If the generating optical pulse is short compared to one cycle of the phonon frequency, the polariton will exhibit a damped oscillation following excitation.

Coherent detection is accomplished by a second femtosecond optical pulse which measures the small change in optical polarizability arising from the electro-optic coupling between the polariton and the electronic oscillator as described by the second driving term in Eq. (2). This approach is an extension of the technique developed by Valdmanis, Mourou, and Gabel[8] for the probing of fast electrical transients in electronic circuits. An important feature of this measurement approach is that it is phase sensitive and produces a signal which is linearly proportional to the polariton amplitude. This makes possible a direct observation of the decay of the polariton and can be used to make coherent measurements of dephasing in real time. An additional feature is that the measurement is nonlocal, i.e., the excitation and probing optical pulses do not overlap spatially. This discriminates against higher-order nonlinear interactions which require overlap, such as the quadratic electro-optic effect, and provides a means of measuring the attenuation and dispersion of the polariton as it propagates over a variable range of distances. It is important to recognize from Eqs. (1) and (2) that the electro-optic measurement is not instantaneous but is also influenced by the

dynamics of the ionic displacement. This introduces an additional damped oscillation into the result similar to the excitation process.

Our specific experimental arrangement is similar to that previously described[7] for generation and detection of far-infrared transients in electro-optic materials. Some important modifications were necessary, however, to observe coherent lattice vibrations. As mentioned, two femtosecond pulses were used: one to generate the radiation field and the other to detect it. Both optical pulses were focused on a sample of lithium tantalate through a common lens and were carefully aligned to propagate parallel through the crystal. The generating pulse was polarized parallel to the c axis of the crystal to produce a radiation field polarized in the same direction.

A unique feature of the use of optical pulses for both generation and detection of the radiation field is the automatic synchronism of the velocities of the radiation field and the probing pulse. The probing pulse "surfs" along the Cherenkov wave front, enabling it to measure the electric field at a stationary point in the wave form by integrating the birefringence along the entire path through the crystal. To plot out the shape of the wave form, the timing of the probing pulse was delayed or advanced relative to the generating pulse by introduction of a variable path length between them.

To make possible the direct observation of the lattice resonance, it was necessary to improve the temporal response of the measurement and also to reduce the effects of far-infrared absorption. This was accomplished by introduction of a modification into our colliding-pulse mode-locked ring laser to control the effects of group-velocity dispersion.[2] This resulted in pulses having a duration of 50 fs (full width at half maximum). Tighter focusing in the lithium tantalate crystal was also used (1.5-μm beam waist) to minimize the time smear due to the finite transverse dimensions of the optical beams. To reduce the effects of infrared absorption, the distance between the generating and detecting beams was reduced to only 7 μm. This was essential to minimize the influence of the strong frequency-dependent absorption near the lattice resonance.

The observed wave form is shown in Fig. 1. Three components contribute to the shape of this wave form. The most prominent feature is the damped ringing on the trailing edge of the wave form following the main pulse. The main pulse arises from the low-frequency response of the electro-optic effect and has been shown previously to consist of approximately one cycle of far infrared, and has a total duration only slightly longer than the optical pulse used for excitation. An additional very rapid transient is evident near the leading edge of the wave form. It arises from the higher-order nonlinearity associated with the quadratic

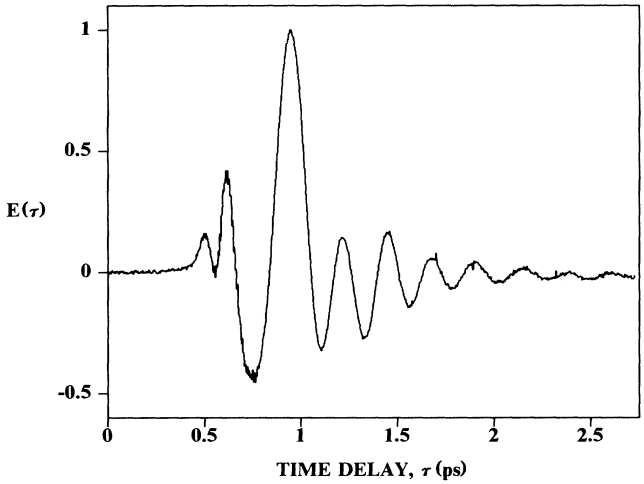

FIG. 1. Polariton wave form in lithium tantalate generated and measured by femtosecond optical pulses with use of the Cherenkov-radiation geometry described in Ref. 7. The damped oscillation on the trailing edge of the main pulse is due to the resonant contribution to the second-order nonlinear optical susceptibility from the TO phonon at 6 THz.

electro-optic effect. Since this term requires spatial overlap of the excitation and probing optical pulses, it can be discriminated against by maintenance of a finite separation between these beams.

The ringing portion of the wave form decays exponentially with a characteristic time of approximately 380 fs. The characteristic frequency of the ringing is approximately 4.2 THz. From detailed measurements of the absorption and dispersion of lithium tantalate, we have previously estimated the TO phonon frequency and damping rate, Γ_i, to be 6.2 and 1.1 THz, respectively.[9] This value of Γ_i corresponds to a phonon dephasing time of 280 fs. The reason for the difference between these values and our experimental results can be understood by a closer examination of the manner in which the lattice resonance contributes to our measurement.

The generation of the polariton wave by optical rectification has a strong resonance due to lattice vibrations, as illustrated by Eq. (1). This resonance has previously been observed by Faust and Henry[10] by difference-frequency generation of optical and far-infrared waves. In the specific case of lithium tantalate, Boyd and Pollack[11] have determined that the electro-optic effect in lithium tantalate is approximately 90% ionic and only 10% electronic in origin. In Fig. 2 we have plotted the expected dispersion of the second-order nonlinear susceptibility, $\chi(\omega)$, as a function of frequency in the low-frequency range near the lattice resonance. This plot is based on the theoretical model of Faust and Henry with use of the specific values for the resonant frequency and damping time of

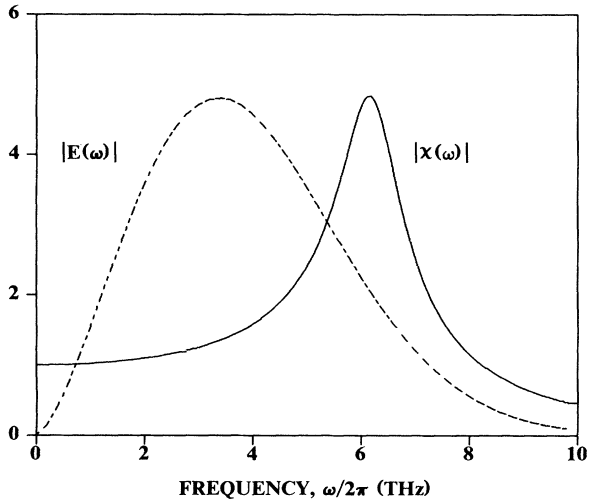

FIG. 2. Dispersion of the second-order nonlinear susceptibility, $\chi_{333}(\omega)$, of lithium tantalate near the TO lattice resonance at 6 THz for the electric field parallel to the polar axis. The magnitude of $\chi_{333}(\omega)$ is plotted here with use of the model of Faust and Henry (Ref. 10) with a damping rate of 1.14 THz determined from far-infrared measurements (Ref. 9) and a value for the ratio of electronic to ionic terms of 0.14 from the work of Boyd and Pollack (Ref. 11). The dashed curve is a plot of the effective spectral response of the measurement technique in the absence of dispersion. An optical pulse width of 50 fs (full width at half maximum), and an optical beam waist of 1.5 μm ($1/e$ radius) are assumed.

the TO resonance which we have previously determined from measurements of the linear absorption and dispersion in lithium tantalate.[9] It shows a peak which rises a factor of 4.8 above the dc value. The width of this peak is determined by the damping rate of the TO phonon. To first order, the resonant properties of the nonlinear and linear susceptibilities are identical. Also, we have assumed that this resonant behavior can be attributed to a single lattice resonance. The strength of the nonlinear susceptibility is proportional to the coupling factor, β_i, in Eq. (1). The spectral amplitude of the polariton wave will be determined by the convolution of $\chi(\omega)$ with the spectral profile of the generating optical pulse in the Cherenkov radiation geometry.[12] The detection process also involves a convolution with $\chi(\omega)$, since the nonlinear susceptibilities for optical rectification and electro-optic detection are the same. This further enhances the resonant features of the experiment.

The propagation of the polariton wave over the finite distance between the generating and detecting optical beams also modifies its shape. Although we have kept this distance extremely small (7 μm), the wave form is nevertheless affected by the strong frequency-dependent absorption and dispersion of the linear dielectric response. The principal effect is to diminish the spectral amplitudes near the resonance and to shift the apparent ringing to a lower frequency.

The combined effects of generation, propagation, and detection of the polariton wave can be estimated more accurately by a numerical simulation of our experiment. The results of this analysis, which will be reported in detail in a later publication, show that the apparent decay time of the lattice vibrations is somewhat greater than the true dephasing time of the phonon. For the particular conditions of our experiment, we expect the observed polariton dephasing time to be only 10% greater than $2/\Gamma_i$. An interesting feature of our experiment is that the apparent resonant frequency of the polariton decreases as the distance between the point of generation and detection increases. From our simulation, we expect the apparent resonant frequency to be lowered from 6.1 to 5.4 THz because of spectral filtering by the linear dielectric response. An important application of our experiment is the determination of the intrinsic speed limit of electro-optic materials. It is clear from our results that the coupling to lattice vibrations, which is an essential ingredient of most practical electro-optic materials, also limits their response time. In the case of $LiTaO_3$ this speed limit is set by the phonon dephasing time and is limited to approximately 380 fs. Other materials having higher phonon frequencies and longer damping times will be faster but will inevitably be less sensitive. Materials such as GaAs, in which the electronic contribution to the electro-optic effect plays a more important role, could overcome this limitation.

We are currently exploring the application of this measurement approach to other materials. A particularly interesting extension of this approach is to the study of nonlinear lattice dynamics. In our current experiment the amplitude of the polariton was approximately 10 V/cm. By use of more intense optical pulses, this amplitude could be increased to the range of 10 kV/cm where the nonlinear response of the lattice could be observed.

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