

Picosecond Acoustic Solitons in Condensed Matter: Generation, Propagation, and Diffraction

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Abstract. Experimental results are presented on propagation and diffraction of high-amplitude, picosecond acoustic wave packets in crystalline solids at liquid-helium temperature. High-intensity femtosecond laser pulses are used to generate these acoustic pulses in a thin-film chromium transducer, yielding a strain of the order of 10^{-3} . Lattice dispersion and anharmonicity determine the nonlinear development, yielding a soliton train of picosecond temporal widths. By reducing the area of excitation from a 1-mm to a 10- μm radius spot, the influence of diffraction can be studied.

INTRODUCTION

Among the most fundamental aspects of solid-state physics, lattice vibrations – or phonons – remain a topic of extensive research. Their dynamics are critical in ultrafast equilibration processes, transport of heat, and material strength. Next to the classical topics, the field of *coherent* phonons has drawn a lot of attention in recent years. The possibility of generating very short (picosecond), *single-cycle*, longitudinal acoustic (LA) phonon wave packets using femtosecond optical excitation of a thin-film metal transducer has led to a range of fundamental and technological applications. Most studies have focused on the propagation of these acoustic pulses in thin multilayer- or nanostructures, using an optical pump- and probe-scheme for detection, at the metal surface, of acoustic reflections inside the structure.

Recently, interest has risen to study nonlinear aspects of propagation, which become apparent after propagation over hundreds of micrometers in the bulk of a crystal [1]. As is known from nonlinear acoustics in fluids, the phenomenon of self-steepening can lead to significant distortion of the initial waveform, eventually resulting in N-wave formation. In the presence of dispersion, the balance of linear- and nonlinear phase accumulations may result in acoustic soliton formation, as was demonstrated earlier in acoustic waveguides [2]. Instead of constructing a dispersion

relation by application of boundary conditions, one can think of using the intrinsic phonon dispersion relation, generated by the lattice itself. For LA phonons, higher-order dispersive terms only come into play for wavevectors significantly far from the center of the Brillouin zone, i.e. for most solids in the THz-frequency region. This would require very short initial wave packets, if not for the presence of self-steepening, which provides with an efficient upconversion mechanism. Recent experiments demonstrate the development of a soliton train from a 50-GHz initial wave packet in sapphire, yielding subpicosecond temporal widths and up to 0.4×10^{-2} acoustic strain amplitudes [3].

EXPERIMENTS

High-intensity acoustic strain pulses are generated using an amplified Ti:sapphire laser setup (Spectra Physics ‘Hurricane’) which provides 130-fs laser pulses at 800 nm with an energy of ~ 0.75 mJ/pulse at a repetition rate of 1.0 kHz. To obtain excitation over a large area, the output of this laser is lightly focused to a spot of several millimeters in diameter onto the sample. The sample is a ~ 10 -mm thick crystal of lead molybdate, with the [001]-direction aligned perpendicular to the surface of incidence. A double-layer consisting of a 500-nm gold film covered with 70 nm of chromium is deposited onto this surface. Small-area, low-amplitude acoustic pulses can be generated using the mode-locked output of the system (Spectra Physics ‘Mai-Tai’), providing ~ 7 nJ/pulse at a repetition rate of 80 MHz. The amplified laser pulses from the Hurricane can also be focused to the same $10\text{-}\mu\text{m}$ waist after severe attenuation, to study the regime of high-intensity, small-area excitation.

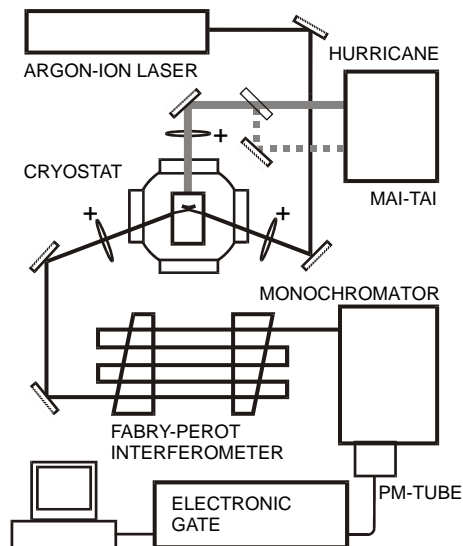


FIGURE 1. Brillouin-scattering setup for detecting high-amplitude acoustic wave packets in a transparent crystal.

For the detection of acoustic strain we make use of the Brillouin-scattering technique. In contrast to time-domain pump and probe methods, which are inherently sensitive only at a reflecting interface, inelastic light scattering enables detection of strain components *in* the bulk of a transparent crystal. Conservation of energy and momentum in the scattering process implies sensitivity to individual Fourier-components of the acoustic wave packet. The setup is shown in Fig. 1. We use a single-mode argon-ion laser operating at 514 nm, focused to a waist of ~ 4 μm in the crystal as an input for the Brillouin-scattering process. The frequency-shifted radiation is analyzed by a quintuple-pass Fabry-Pérot interferometer and standard photon counting setup. The short time window of several nanoseconds for interaction with the travelling acoustic pulse in the scattering volume demands for electronic gating of the

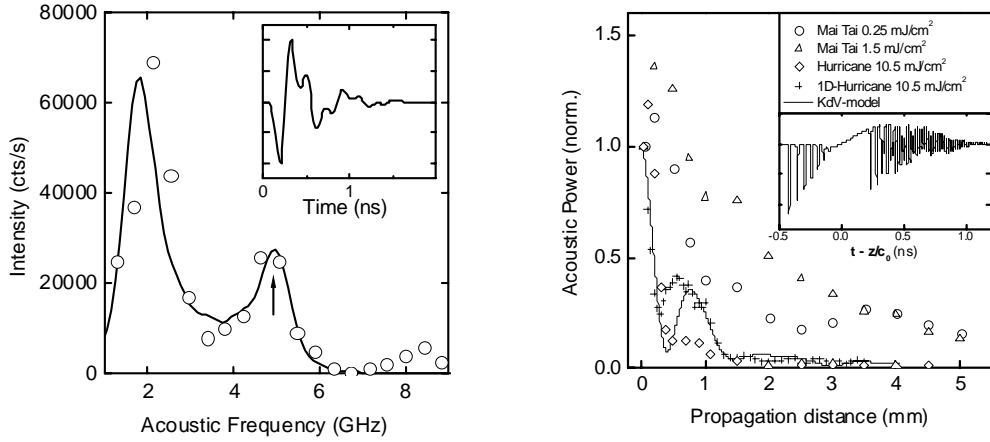


FIGURE 2. (left) Acoustic power spectrum. Points: experimental data. Line: Power spectrum obtained from waveform shown in inset. Arrow indicates frequency where further experiments have been performed. (right) Propagation of the 5-GHz spectral component for: small-area excitation ($w_0=10\ \mu\text{m}$), for three pump-laser fluence values, and large-area excitation at $10.5\ \text{mJ}/\text{cm}^2$. Line: simulation of KdV-equation. (inset): wave train at 2.0 mm resulting from simulation.

signal when the low repetition-rate pump laser is used. Scans of the phonon-beam profile perpendicular to the direction of propagation can be made with micrometer resolution by tilting the pump beam using a galvanometer-mounted mirror (Cambridge Technology Model 6210).

Fig. 2 (left) shows a typical power spectrum of the initial acoustic wave packet in the crystal, after a $100\text{-}\mu\text{m}$ travelling distance, obtained by measuring the Brillouin intensity as a function of scattering angle. The spectrum can be fitted to reasonable agreement with a waveform consisting of multiple reflections of $\sim 45\%$ from the gold-lead molybdate interface and less than $\sim 15\%$ reflection from the chromium-gold interface. The large temporal width of the pulse implies that soliton formation will only start up after a significant amount of self-steepening of the packet.

In the following we will concentrate on a single frequency-component of the wave packet, which we have chosen to be the sub-maximum at 5 GHz. The propagation and diffraction of this component for a small-area initial packet was followed through the crystal at three settings of the pump-laser fluence, covering the entire dynamic range of our experiment. The peak amplitude of these profiles is shown in the right part of Fig. 2. At the highest pump fluence under study, the evolution of a large-area, nondiffracting packet was also measured for comparison. It is seen that there is a significant difference in evolution at the three pump fluences under study. We know from simulations based upon the KdV-equation that the initial decay of the power at low frequencies can be attributed to self-steepening of the wave packet. It is not surprising that this effect is less pronounced at lower pump intensity. Some features, like the initial *increase* of the peak power at 0.25 and $1.5\ \text{mJ}/\text{cm}^2$, and the fact that the $0.25\ \text{mJ}/\text{cm}^2$ trace decreases more rapidly than that at $1.5\ \text{mJ}/\text{cm}^2$, are not yet well understood, although the latter may be due to differences in diffraction losses. Comparison of the two traces at $10\ \text{mJ}/\text{cm}^2$ shows a faster decrease of peak intensity

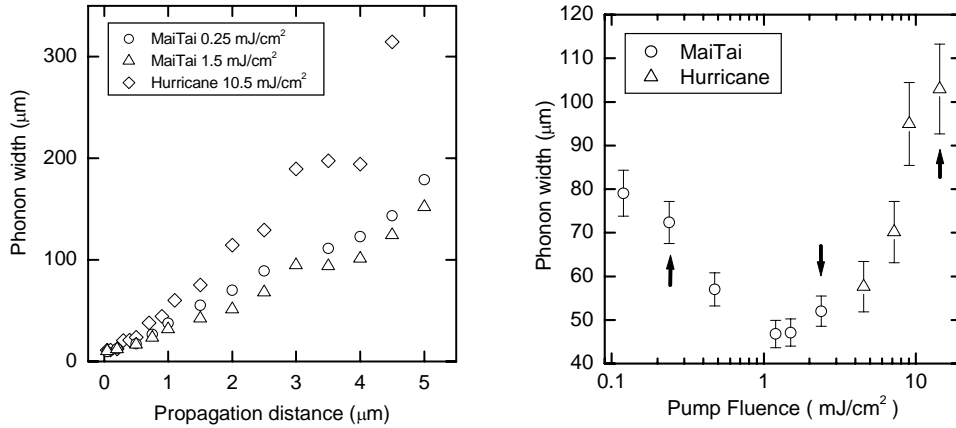


FIGURE 2. (left) Divergence of the 5-GHz frequency component of the acoustic wave packet for different pump fluences, at a temperature of 5 K, under small-area excitation. (right) Beam 1/e half width at 2.0 mm propagation distance. Arrows indicate values where traces have been taken.

for the “small-area” trace, followed by a oscillation of lower amplitude than that in the “large-area” curve. Although the faster decay could indicate a speeding-up of the self-steepening process, the combination with the decrease of the oscillation amplitude makes us believe that part of the initial spectral power is diffracting away from the central part of the beam. Evidence of this can be found in the left part of Fig. 3, which shows the diffraction of the three small-area traces. We observe an increase in beam width of almost an order of magnitude within the first millimeters of propagation. Further, the diffraction is seen to depend on pump fluence, as is made even more clear in the right part of Fig. 3. We should keep in mind that the wave packet consists of different polarities, each responding in a different way to the combined nonlinearity, dispersion, and diffraction effects. As all three processes are of the same order of magnitude in this experiment, we need to consider the full, 3-dimensional, propagation of these wave packets []. We will attempt to explain this behaviour in the near future.

References

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