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Propagation of ultrashort acoustic wave packets in $PbMoO_4$ studied by Brillouin spectroscopy

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Abstract

We employ Brillouin spectroscopy to study the propagation in $PbMoO_4$ of ultrashort acoustic wave packets created by absorption of subpicosecond optical pulses in a thin-film metallic transducer. The development of the acoustic power spectrum can be studied up to several tens of GHz. We obtain information on the local propagation velocity with a sensitivity of 0.1%. Diffraction of the Fourier components of the acoustic wave packet in the crystal can be monitored in the spectral domain via the spectral width, or in real-space using scans transverse to the beam, and turns out to be anomalously small. Preliminary results at helium temperatures are presented, which show pump intensity dependent propagation effects. © 2002 Elsevier Science B.V. All rights reserved.

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The research area of picosecond ultrasonics has received an increasing amount of interest in both fundamental and industrial research during the last decade [1–3]. Most studies focus on thin-film structures covered with a metallic transducer, where the optical skin depth limits the time resolution of the experiment. Only very recently, explorations emerge to examine the propagation of picosecond acoustic pulses in bulk crystalline materials [1], although a metallic transducer remains necessary for probing the development of the wave packet. Frequency domain techniques have been used to study low amplitude, monochromatic coherent phonons [4]. We demonstrate that ultrashort, broadband acoustic pulses can be studied in the bulk of a transparent crystal by means of frequency domain techniques.

We use Brillouin scattering to monitor the different frequency components as they develop during the propagation of the acoustic wave packet. For an introduction to the principles of Brillouin scattering we refer to the existing literature [5]. A plane acoustic wave packet reflects a minute fraction of an incoming laser beam, which for anti-Stokes Brillouin scattering is Doppler shifted upward, and for Stokes shifted downward, by the phonon frequency corresponding to the scattering wave vector. This implies that for each scattering angle we are sensitive to a single Fourier component of the acoustic wave packet. The condition for wave vector conservation is relaxed by some amount due to the limited size of the interaction volume, determined by the spatial overlap of the focused Brillouin laser beam and the Fourier component of the acoustic pencil. As a consequence, the observed spectral width of the Brillouin signal contains direct information on the

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transverse width of the acoustic beam for the selected frequency component, provided that the opening angle of the detection setup is sufficiently large.

The sample is a $10 \times 4 \times 5\,\mathrm{mm}^3$ single crystal of $PbMoO_4$ with its *c*-axis perpendicular to the $10 \times 4\,\mathrm{mm}^2$ surface. A 600 nm thin gold transducer is deposited onto one of these surfaces, with a 5 nm intermediate layer of Cr for better adhesion. Lead molybdate is a widely used medium for acousto-optic modulators and has been chosen by us mainly for its very high acousto-optic coupling parameter. Given the sound velocity in the [0 0 1] direction of 3.64 km/s and the index of refraction of 2.26, the maximum frequency that can be detected by Brillouin scattering is limited to 32 GHz at a laser wavelength of 514 nm.

Acoustic wave packets are created by absorption of light in the gold film from a mode-locked Ti–sapphire femtosecond laser, focused to a Gaussian spot with a waist of 12 μm. The length of the acoustic pulse created in a metallic film is determined by the nonequilibrium electron transport distance, reached within the relaxation time of a few picoseconds, and amounts in gold to, say, 150 nm [6]. We use two excitation geometries, by focusing the pump beam at the crystal–gold interface (I) or at the air–gold interface (II). Both configurations produce ambipolar pulses with equally sharp features, but configuration I contains significantly more acoustic power in the lower frequency components of the wave packet.

The local acoustic strain for the selected Fourier components is detected by Brillouin scattering employing a single-mode argon-ion laser operating at 514 nm. We focus the laser beam in the crystal to a measured waist of 4.5 μm. The frequency-shifted radiation is analyzed by a quintuple-pass Fabry–Perot interferometer (Burleigh RC110) and detected by standard photon counting techniques. The spectrometer has been used earlier to study the propagation of coherent, monochromatic phonon beams in lead molybdate and paratellurite [4].

A typical Brillouin spectrum of the acoustic wave packet in configuration I, at a scattering angle of 20°, is presented in Fig. 1a. The spectrum consists of a series of sharp, finesse-limited, peaks at multiples of the 75.4 MHz repetition frequency of the Ti–sapphire laser, centered around a Doppler shifted frequency of 3.5 GHz. The mode structure in the spectrum is due to optical interference inside the interferometer of subsequent acoustic pulses traversing the interaction volume. The phase relationship between the light scattered from subsequent acoustic pulses implies that we are dealing with a mode-locked acoustic pulse train. We checked that the spectral width is determined by the width of the acoustic beam in the scattering plane by measuring at different diameters of the Ti–sapphire beam at the transducer.

The signal from excitation geometry II was found to be a factor of 40 weaker than that of I under the same conditions. We attribute this to a strong difference in the acoustic power in the selected frequency component. We checked that the dependence of the Brillouin intensity on

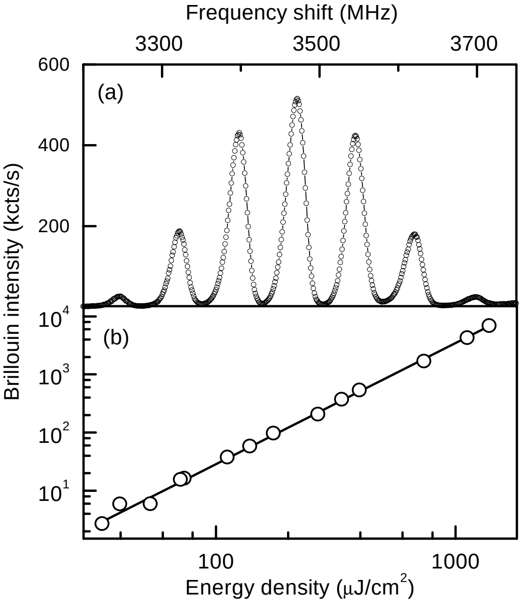

Fig. 1. (a) Brillouin spectrum observed at a scattering angle of 20°, showing a Doppler shifted envelope consisting of peaks with 75.4 MHz mutual distance. Width of the peaks is limited by the resolution of the spectrometer. (b) Dependence of the integrated Brillouin intensity, measured at 100 μm distance from the transducer, on excitation power, showing a quadratic dependence over 2 orders of magnitude.

propagation distance is similar in both configurations. Due to the low intensity of the signal in configuration II, we will limit ourselves in the following to results from configuration I.

The dependence of the Brillouin intensity on excitation power is quadratic, as can be seen in Fig. 1b. This suggests that the strain generation in the gold film in the measured frequency range is linear. Comparison of the measured Brillouin intensity to the thermal background of LA phonons leads to a calculated "temperature" of the acoustic mode exceeding 10^6 K around 3.5 GHz.

To obtain information on the propagation of the selected acoustic mode in the crystal, we measured the acoustic beam profile as a function of propagation distance. This was done by scanning of the acoustic pencil through the detection volume in the vertical direction. The measured profiles, shown in Fig. 2a, are of Gaussian shape and show broadening due to Fraunhofer diffraction. The obtained divergence angle is surprisingly small and does not correspond to experimental results on low amplitude monochromatic phonons of the same frequency that do match the theoretically predicted value [4]. Integration of the signal gives the total acoustic power, which is shown versus propagation distance in Fig. 2b. The decay can be fit by a single exponential function with a relaxation length of 225 μm, which is in agreement with the theory for thermal damping [4].

Experiments at liquid helium temperatures are necessary to eliminate thermal damping, thus enabling the study of propagation over much longer distances and at lower pump intensities. Preliminary results indicate a strong dependence of both the divergence (not shown) and the acoustic power (inset of Fig. 2b) of the 3.5 GHz frequency component on the pump intensity. The rapid decay of the total intensity at the highest pump powers may be explained by an increase in coupling between acoustic modes, due to the high local strain around the wave packet maximum. We observed this population decay to be even more pronounced at lower acoustic frequencies, around 1 GHz, suggesting the presence of an upconversion effect, which may be interpreted as the self-steepening of the acoustic wave packet. The beam divergence was observed to decrease for increasing pump intensities, for which we cannot give a satisfying explanation at this moment.

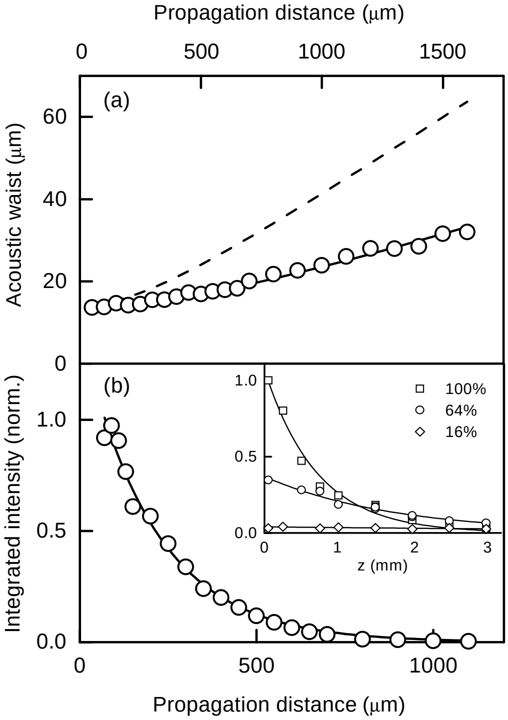

Fig. 2. (a) Acoustic beam width at 3.5 GHz as a function of propagation distance in the crystal at room temperature. Full line: theoretical fit to data corresponding to Fraunhofer diffraction combined with weak phonon (de)focussing. The resulting frequency corresponding to the fit is 7.5 GHz, which is a factor of 2.2 too high. Dashed line: predicted divergence at 3.5 GHz. (b) Integrated Brillouin intensity at 3.5 GHz as a function of propagation distance at room temperature. Line: theoretical fit to data corresponding to single exponential relaxation, $l_{\mathrm{ph}} = 225\,\mu\mathrm{m}$, in agreement with the thermal damping length at 3.5 GHz [4]. Inset: Brillouin intensity as a function of propagation distance z at 15 K for three different pump intensities (100% equals 1100 μJ/cm^2 pump intensity).

Further investigation into the power dependence of these effects and of diffraction are in progress. Also experiments at frequency components up to 30 GHz are planned in the near future. Nonlinear acoustic effects—if present—are expected to be more pronounced at higher frequencies.

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References

[1] H. Hao, H.J. Maris, Phys. Rev. Lett. 84 (2000) 5556.

[2] T.F. Crimmins, A.A. Maznev, K.A. Nelson, Appl. Phys. Lett. 74 (9) (1999) 1344.

[3] D.H. Hurley, O.B. Wright, Opt. Lett. 24 (18) (1999) 1305.

[4] E.P.N. Damen, A.F.M. Arts, H.W. de Wijn, Phys. Rev. Lett. 74 (1995) 4249;
E.P.N. Damen, A.F.M. Arts, H.W. de Wijn, Phys. Rev. B 59 (1999) 349.

[5] J.W. Tucker, V.W. Rampton, Microwave Ultrasonics in Solid State Physics, North-Holland, Amsterdam, 1972.

[6] G. Tas, H.J. Maris, Phys. Rev. B 49 (1994) 15046.