

Trains of ultrashort acoustic solitons

Otto L. Muskens* and Jaap I. Dijkhuis**

Debye Institute, Department of Physics and Astronomy, University of Utrecht, P.O. Box 80 000, 3508 TA, Utrecht, The Netherlands

Received 13 July 2004, revised 20 September 2004, accepted 25 October 2004

Published online 22 November 2004

PACS 43.25.Rq, 62.30.+d, 63.20.-e

We present results on soliton trains in a PbMoO_4 crystal, and show that a wide range of soliton train parameters is accessible using different transducer materials. Brillouin spectra are presented probing the development of high-amplitude bipolar strain pulses into soliton trains during propagation in PbMoO_4 at liquid helium temperatures. A simple interpretation of the beating patterns in the Brillouin scattering yields the highest relative soliton velocities in the trains of up to 0.3%. We use this to predict a value for the third-order elastic constant of PbMoO_4 of $C_{333} = -480$ GPa. The extremely fast solitons predict participation of the entire LA phonon branch in the soliton formation. For a gold transducer, the solitons are much wider but their number in the train is even larger.

© 2004 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

1 Introduction

The technique of picosecond ultrasonics has recently been extended to injecting longitudinal, bipolar strain pulses with exceptionally high amplitudes. In that regime, the acoustic nonlinearity leads to self-steepening of the propagating strain pulse in crystals and at room temperature to the formation of a so called damped N wave [1]. This self-steepening process can be directly monitored via the reduction and subsequent oscillations of the Brillouin light scattering signal probing the low-frequency components of the strain pulse. At liquid helium temperatures, damping is virtually absent and the generation of higher frequency components is only limited by the inherent dispersion of crystals. This ultimately leads to the development of solitons as was demonstrated for the first time in a pump–probe experiment by Hao and Maris [2]. Using much broader acoustic beams and higher pump fluences, we showed the development of soliton trains in sapphire via a typical beating pattern in the Brillouin scattering intensity [1, 3]. Traces computed using the Korteweg–de Vries equation and the known parameters of sapphire appeared to produce patterns that faithfully track all experimental data. From these extensive simulations, it appeared that the time duration of typical solitons in the train can reach values as short as 0.5 ps. Recently, we demonstrated that solitons are capable to impulsively excite THz-crystal field transitions of chromium impurity ions in ruby [4].

In this paper we explore Brillouin scattering signals on developing and propagating soliton trains in the much softer crystal PbMoO_4 and compare the results to the case of sapphire for identical input strain amplitudes. Without the need of carrying out extensive computer simulations, we obtain directly from the experiment a value for the unknown nonlinear elastic constant of PbMoO_4 . Our results using chromium transducers further predict that solitons have widths approaching the lattice parameter in PbMoO_4 .

* Current address: CPMOH, Université Bordeaux 1, 351 Cours de la Libération, 33405 Talence, France

** Corresponding author: e-mail: J.I.Dijkhuis@phys.uu.nl, Phone: +3130 253 33 02, Fax: +31 30 253 74 68

Table 1 Comparison of relevant physical parameters of the two crystals under study.

	c_0 (10^3 m/s)	ρ (10^3 kg/m ³)	C_{33} (GPa)	C_{333} (GPa)	β (10^{-17} m ³ /s)
Al ₂ O ₃ [0001]	11.23	3.98	502	−3340	3.5
PbMoO ₄ [001]	3.64	6.94	92	− 480 [†]	2 [‡]

[†] Value estimated from the experiments in this paper.

[‡] Value estimated from the LA-phonon dispersion curve.

and that the train may carry up to 24 solitons at feasible excitation powers. In case of gold transducers, producing much longer input strain pulses, the train develops slower but contains even more solitons.

2 Trains of solitons

In the following we demonstrate how Brillouin scattering can be used to obtain direct quantitative estimates on soliton trains in a crystal, without the need of extensive numerical simulations. We compare the results for a sapphire (Al₂O₃) single crystal covered with a chromium transducer, as used earlier in Refs. [1, 3], with new Brillouin traces measured in a lead molybdate (PbMoO₄) crystal with either a chromium or a gold transducer. Lead molybdate is quite convenient because of its exceptionally high elasto-optic coupling coefficient and large soliton Brillouin signals.

This choice of crystals covers a wide range of acoustic properties. Table 1 shows the large difference of the mass density ρ and the elastic constant C_{33} , resulting in a factor of 3 difference in the velocity of longitudinal acoustic phonons c_0 . The higher-order dispersion constant β may be estimated from the cutoff of the LA-phonon branch at about 100 cm^{−1}. Although it is known that this may overestimate the dispersion by almost an order of magnitude [5], we use this value in the case of PbMoO₄ (cf. Table 1) to provide an upper limit to the soliton width and a lower limit to the number of solitons in the train. The amplitude and velocity of the solitons are however not influenced by this constant but depend only on the initial strain and the nonlinearity parameter. Unfortunately, the nonlinear material constants of the PbMoO₄ crystal are not known precisely. We will show, however, that an estimate for the third order elastic constant C_{333} can be directly deduced from the soliton velocities obtained from the Brillouin experiment.

First, let us consider the case of a PbMoO₄ crystal covered with a chromium transducer. As the thermoelastic generation mechanism of the picosecond strain pulse depends exclusively on the metal film, we may safely assume that the initial wavepacket contains the same frequency components as in the Al₂O₃/chromium case described in detail earlier [1]. An important difference however is the much higher acoustic reflection coefficient at the interface between transducer and crystal. For the PbMoO₄ crystal this reflection amounts to about 60% of the strain amplitude of the transmitted pulse [3]. However, the transmitted strain in the crystal is enhanced by the compression due to the smaller sound velocity in PbMoO₄ by a factor c_{tr}/c_0 , where c_{tr} denotes the velocity in the transducer (cf. Table 2). The net effect

Table 2 Properties relevant for soliton trains of three crystal/transducer combinations: width of gaussian derivative strain profile τ_0 , strain reflection coefficient r at transducer–crystal interface, sound velocity ratio c_{tr}/c_0 determining the strain enhancement in crystal. Calculated relative velocity of first soliton v_{sol}/c_0 , shock formation distance z_{shock} , and number of solitons in train N , for a typical initial strain s_0 of 10^{-3} .

	τ_0 (ps)	r	c_{tr}/c_0	$10^3 v_{sol}/c_0$ [†]	z_{shock} (μm) [†]	N [†]
a) Al ₂ O ₃ /chromium	7.0	0.2	0.5	1.2	65.5	10
b) PbMoO ₄ /chromium	7.0	−0.6	1.7	0.6	42.5	2 [†]
c) PbMoO ₄ /gold	80.0	0.3	0.9	0.6	485.0	24 [‡]

[†] Calculated for $s_0 = 10^{-3}$.

[‡] Calculated for $\beta = 2 \times 10^{-17}$ m³/s.

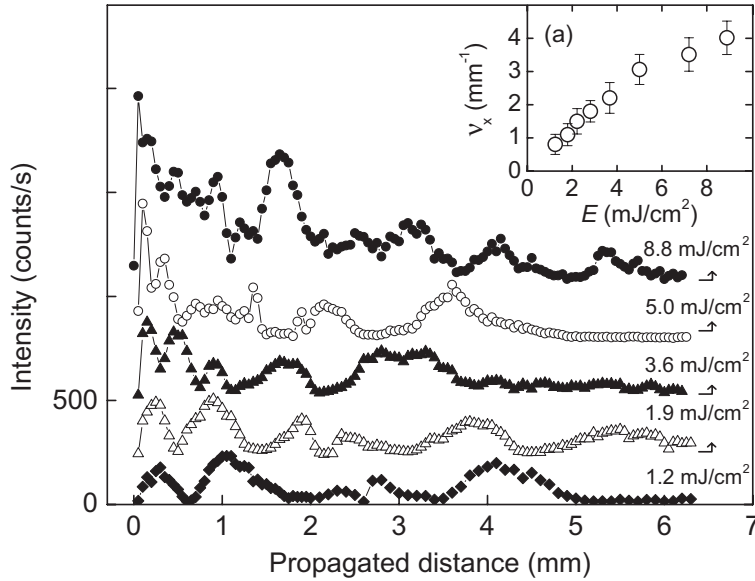


Fig. 1 Brillouin traces at 5 GHz in PbMoO_4 [001] with a chromium transducer, at a temperature of 5 K. Arrows indicate vertical offsets, numerical values denote pump fluence E . (a) Maximum spatial frequency ν_x against E at a Frequency of 5 GHz.

is a factor-of-two increase in strain amplitude for PbMoO_4 compared to Al_2O_3 , at identical pump intensities. For example, a typical pump fluence E of 8.5 mJ/cm^2 leading to an acoustic strain amplitude of the injected strain pulse in the sapphire crystal of 2×10^{-3} , will produce a strain of 4×10^{-3} in the PbMoO_4 .

Figure 1 shows the Brillouin intensity as a function of propagation distance of an intense strain pulse in a PbMoO_4 crystal measured at an acoustic frequency ν_B of 5 GHz, at low temperature. We observe an initial rise and decay of the Brillouin intensity that becomes extremely sharp at high pump intensities, signifying an increasingly strong self-steepening effect of the acoustic wavepacket. Subsequently, a complicated oscillation pattern is observed that is characteristic for soliton development and has been demonstrated to correspond to spatial resonances and Bragg interference of the light scattered by the moving soliton train [1, 3]. The period of the fastest oscillation in this pattern corresponds to the interference of light scattered from the fastest soliton in the train with the light reflected from the slowest solitons and the linearly propagating parts of the wavepacket. This period, shown as the spatial frequency ν_x against pump fluence in Fig. 1(a), allows us to very accurately determine the soliton velocity ν_{sol} in the moving frame, i.e. relative to that of linear phonons, by the formula [3]

$$\nu_{\text{sol}} = \frac{\nu_x c_0^2}{\nu_B} \quad (1)$$

For the highest beating period of 4 mm^{-1} , this results in a relative velocity increase of the soliton of ν_{sol}/c_0 of 3×10^{-3} . Further, we know that the amplitude of a soliton is directly related to its velocity via the relation

$$a_{\text{sol}} = \frac{6\rho c_0 \nu_{\text{sol}}}{\alpha} \quad (2)$$

where $\alpha = C_{333} + 3C_{33}$ is the nonlinear constant emerging in the acoustic wave equation and $a_{\text{sol}} = 2s_0$ denotes the strain amplitude of the soliton. Given the above estimate of the initial strain s_0 of -4×10^{-3} , we arrive at an estimate for the third-order elasticity parameter of PbMoO_4 of $C_{333} \approx -480 \text{ GPa}$.

Using this value for the nonlinear constant and our rough estimate of the phonon dispersion constant β , we can determine the spatial width of the solitons and the expected number of soliton pulses in the train for a given input strain pulse. For this we define the similarity parameter σ familiar from earlier

treatments of the Korteweg–de Vries solitons [6]. Expressed in the material constants and the initial strain pulse this parameter reads

$$\sigma = 0.50 c_0 \tau_0 \left(\frac{\alpha s_0}{12 \rho c_0 \beta} \right)^{1/2}, \quad (3)$$

where the numerical multiplication factor follows from our definition of the pulse width τ_0 [3]. The number of solitons in the train can readily be estimated at large σ using $N \sim \sigma$, and the width of the n -th soliton is found from $\tau_n = \tau_0 (2s_0/a_n \sigma^2)^{1/2}$, with a_n the soliton amplitude. For the above experimental configuration, we arrive at a number $N \sim 7$ and width $\tau_1 = 0.94$ ps, taking the the upper estimate of $\beta = 2 \times 10^{-17}$ m³/s. For a ten times smaller value of β , which is more realistic given the experimental results for other crystals, the number of solitons in the train increases more than threefold to an astonishing $N \sim 24$ and soliton width as small as 0.29 ps. The spatial widths of the individual solitons ranges between 1.0 and 3.0 nm for the two dispersion values, close to the lattice parameter of PbMoO₄ ($c = 12.11$ Å).

Clearly the experiment in the PbMoO₄/chromium sample reaches the physical limits of the material both in strain amplitude as in spatial extent of the soliton pulses. Consequently, the nonlinear development of the acoustic wavepacket virtually needs the full LA-phonon branch, and the weak dispersion approximation is no longer valid. We failed to detect deviations in the soliton train development due to these higher-order nonlinear and dispersive effects expected at large propagation distances, because we encountered difficulties in reproducing the Brillouin experimental traces beyond the first few oscillation periods. The complicated pattern in the Brillouin intensity appeared to be much more sensitive to changes in the alignment of the system and to the position of the pump on the transducer compared with our earlier measurements in sapphire.

As a second example, we present an investigation of the development of strain pulses in a PbMoO₄ sample covered with a 500 nm gold film. As the electron–phonon coupling in gold is much weaker than in chromium, the width of the injected strain pulses is an order of magnitude larger, as shown in Table 1, and correspondingly the acoustic frequency components are much lower. Figure 2(a) shows the meas-

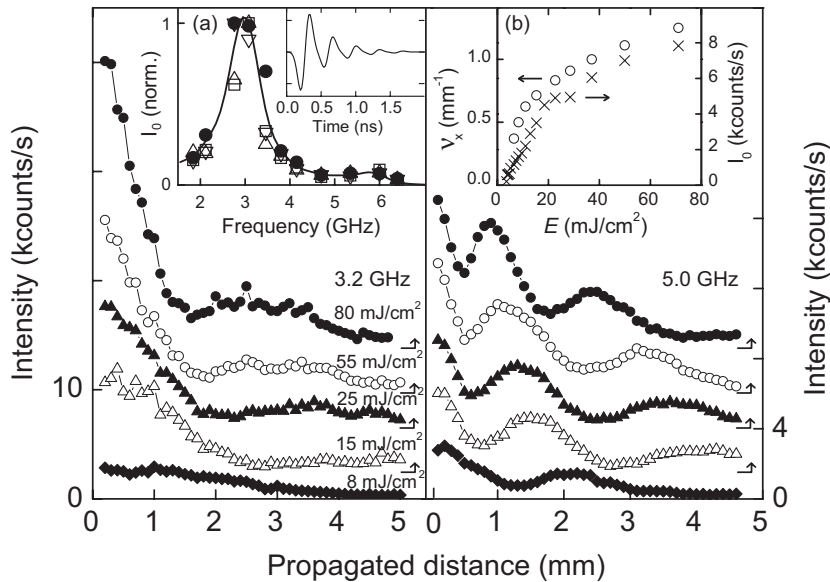


Fig. 2 Brillouin traces at 3.2 GHz (left) and at 5.0 GHz (right) in PbMoO₄ [1] with a gold transducer, at a temperature of 5 K. Arrows indicate vertical offsets, numerical values denote pump fluence E . (a) Acoustic power spectrum of the strain waveform, (dots) experimental data, (line) fit assuming a strain waveform given by (inset). (b) Maximum spatial frequency ν_x (\circ , left scale) and Brillouin intensity at $z = 50$ μm , I_0 (\times , right scale), against E for the 5 GHz frequency component.

ured Brillouin intensity as a function of scattering angle, yielding the frequency spectrum of the acoustic wavepackets. This power spectrum can be fit to good agreement assuming the usual bipolar waveform [7] with a reflection at the crystal interface of about 30% [inset of Fig. 2(a)]. As the absorption of gold at the optical pump wavelength (800 nm) is much less (about 2%) than than of chromium (about 60%), the applied pump fluences can be taken an order of magnitude higher without damaging the film.

The development of two typical frequency components, namely at the peak (3.2 GHz) and beyond (5.0 GHz), is monitored as a function of propagated distance (see Fig. 2). Clearly, the peak at 3.2 GHz decreases at all pump fluences over a distance of one to several millimeters, indicating that the self-steepening is a slow process for this wide initial strain pulse. For an initial strain amplitude of order 10^{-4} , the peak of the strain pulse overtakes the wavefront over a propagated distance $z_{\text{shock}} \approx c_0^2 \tau / v_{\text{sol}}$ that amounts to 5 mm. This corresponds well with the decay of the spectral peak at 3.2 GHz for lower pump fluences.

The higher frequency component at 5.0 GHz shows a typical oscillation that can again be analyzed in terms of a spatial beating of parts of the wavepacket moving at different velocities. Figure 2(b) shows the resulting spatial frequencies ν_x against pump fluence E . For comparison, also the Brillouin intensity of this frequency component, taken at the position of the transducer, is shown. Clearly, the two correlate well, implying that the oscillation period is proportional to the strain amplitude, as was shown earlier in Fig. 1(a). The highest value of ν_x of 1.3 mm^{-1} corresponds to a relative soliton velocity increase, according to Eq. (1), of 9.5×10^{-4} , or an initial strain s_0 , according to Eq. (2), of 1.3×10^{-3} . This yields a value of the number of solitons in the train of $N \sim 30$ and a soliton width of 2.7 ps, for the highest estimate of the dispersion β . Of course these numbers depend on the choice of β . Clearly the development of strain packets in the gold film covers a completely different regime than the soliton trains generated using the picosecond pulses from the chromium transducer. The soliton pulses have lower amplitude, are broader but the train consists of a larger number of solitons.

3 Conclusions

We have studied employing Brillouin spectroscopy acoustic soliton trains in PbMoO_4 and compared the results to the ones in the much harder material sapphire. We found in case of using chromium transducers, that the width of the individual solitons approaches the lattice constant. This implies that virtually the full longitudinal acoustic phonon branch participates in the soliton and that higher-order corrections to the dispersion are needed. In case of gold transducers the trains need more time to develop and the solitons are wider, but the number of solitons in the trains may exceed 30. This all may serve as an illustration for the wide range of soliton trains that can be produced using high-power excitation of metal transducers.

Acknowledgement The authors wish to thank P. Jurrius and C. R. de Kok for their technical assistance. This work was supported by the Netherlands Foundation “Fundamenteel Onderzoek der Materie (FOM)” and the “Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO)”.

References

- [1] O. L. Muskens and J. I. Dijkhuis, Phys. Rev. Lett. **89**, 285504 (2002).
- [2] H.-Y. Hao and H. J. Maris, Phys. Rev. B **64**, 064302 (2001).
- [3] O. L. Muskens and J. I. Dijkhuis, Phys. Rev. B **70**, 104301 (2004).
- [4] O. L. Muskens, A. V. Akimov, and J. I. Dijkhuis, Phys. Rev. Lett. **92**, 035503 (2004).
- [5] H.-Y. Hao and H. J. Maris, Phys. Rev. B **63**, 224301 (2001).
- [6] G. B. Whitham, Linear and Nonlinear Waves (Wiley-Interscience, New York, 1974).
- [7] D. H. Hurley and O. B. Wright, Opt. Lett. **24**(18), 1305 (1999).