



The 29-cm^{-1} ruby phonon detector as a probe for ultrashort strain solitons

O.L. Muskens^{a,*}, S. Purushothaman^a, A.V. Akimov^{a,b}, J.I. Dijkhuis^a

^aFaculty of Physics and Astronomy, Debye Institute, Ornstein Laboratory, Atom Optics and Ultrafast Dynamics, University of Utrecht, P.O. Box 80 000, 3508 TA, Utrecht, Netherlands

^bIoffe Physico-Technical Institute, Russian Academy of Sciences, Politechnicheskaya 26, St. Petersburg 194021, Russia

Abstract

We demonstrate the interaction between ultrashort acoustic strain solitons and the 29-cm^{-1} electronic two-level systems in photoexcited ruby. For propagation along the crystallographic c -axis we observe a strong soliton-induced electronic population only after reflection at the far end of the crystal. We attribute this to mode-conversion and diffuse scattering at the crystal surface, which breaks the inhibited coupling of the soliton travelling along the c -axis. Model calculations show good agreement assuming a hemispherical scattering source and dipolar form for the interaction strength.

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1. Introduction

Recent experiments have demonstrated the development of ultrashort strain solitons from a picosecond acoustic wavepacket during ballistic propagation in large crystals at low temperatures [1,2]. It was found that these *half*-cycle pulses can reach strain amplitudes of 0.4% and frequency components into the THz-range. This combination of high strains on ultrashort time scales holds promise for exploring THz-coherent electron–phonon interactions in a medium containing two-level centers [3,4]. Early experiments have

explored the phenomenon of acoustic self-induced transparency of GHz ultrasonic pulses [5], demonstrating the direct analogy with coherent optics in two-level atoms [6]. For THz electron–phonon interactions, however, coherence has up to now not played a significant role, mainly due to the lack of suitable excitation mechanisms for the strain field. Stimulated emission [7] has been well described by incoherent rate equations, and it was only in phonon-induced optical dephasing experiments [8] that the question of phonon coherence was addressed.

In this paper, we make use of the well-known 29-cm^{-1} phonon spectrometer based on the $\bar{E}(^2E) - 2\bar{A}(^2E)$ transition in optically excited ruby. By coupling this system to the ultrashort soliton train of Ref. [2], we have already

*Corresponding author. Tel.: +31-30-253-2206; fax: +31-30-253-7468.

E-mail address: o.l.muskens@phys.uu.nl (O.L. Muskens).

demonstrated coherent, impulsive electron–phonon interactions along the crystallographic a -axis [3]. Here, we extend the studies to propagation in the high-symmetry direction, the c -axis.

2. Results and discussion

High-amplitude, picosecond strain wavepackets are generated by ultrafast excitation of a thin chromium film using mJ optical pulses from an amplified Ti:sapphire laser, operating at 1 kHz [2]. The sample is a 500 ppm. Cr ruby crystal of $9.7 \times 10 \times 15$ -mm³ dimensions, its c -axis oriented perpendicular to the 10×15 -mm² surface. A 100-nm chromium film is deposited onto this surface. The sample is immersed in superfluid helium, and pumped to a temperature of 1.5 K. A small magnetic field generated by a split-coil magnet is used to lift the degeneracy of the Kramers doublets, speeding up the detector response by a factor of 4 [9].

The metastable $\bar{E}(^2E)$ -level is populated by indirect excitation through the higher absorption bands, using a 2-W argon-ion laser focused to a pencil of about 200 μm in diameter. During the propagation of the strain pulses, the time evolution of the R_1 - and R_2 -emission lines is monitored using a double monochromator equipped with a time-resolved photon counting setup with a resolution of 3 ns. Direct excitation of the $2\bar{A}(^2E)$ -level during the acquisition time is suppressed by switching off the laser beam using an AOM. Results for propagation along the ruby a -axis using the same experimental configuration [3] showed strong interaction between strain solitons and the electronic system through the whole crystal. In the current experiment along the c -axis, however, no soliton-induced R_2 -luminescence could be observed inside the crystal.

Similar suppression of the LA phonon-induced R_2 luminescence along the c -axis was observed and explained in collimated heat pulse experiments by Kaplyanskii et al. [10]. By applying the selection rules for the $\bar{E}(^2E) - 2\bar{A}(^2E)$ transition in the trigonal point group, they demonstrated complete disappearance of the electron–phonon coupling parameter for propagation along the c -axis.

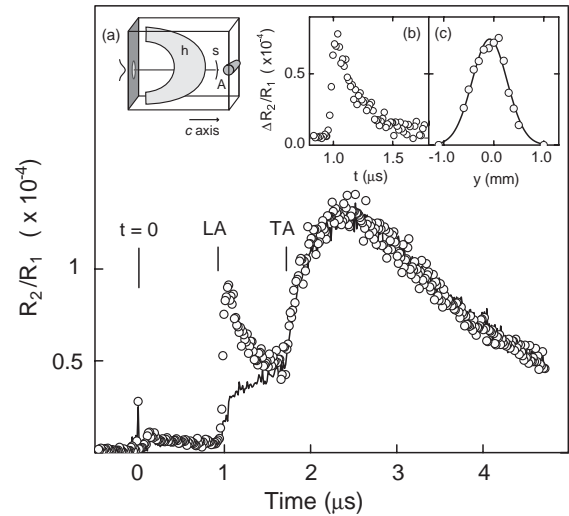


Fig. 1. Time-resolved R_2 -luminescence normalized to R_1 at 9.6-mm propagation distance along the c -axis, for on-axis (\circ) and off-axis (line) configurations. (a) Experimental configuration, with A detection region, s soliton pulse and h heat pulse. (b) Difference signal between on- and off-axis. (c) Transverse profile of the soliton-induced LA intensity at $z = 9.6$ mm.

Strikingly, however, we do observe a soliton-induced contribution at the far end of the crystal. Fig. 1 shows two typical time-resolved luminescence traces, for the configurations in which the excited zone A (c.f. Fig. 1(a)) is placed in the path of the acoustic wavepacket (on-axis) or at a transverse displacement of 1 mm (off-axis). A difference is observed at a time corresponding to the arrival of LA-phonons from the transducer. Fig. 1(b) shows the difference of the two traces around the LA arrival time. We observe a peak followed by an exponential tail of $\sim 0.25 \mu\text{s}$ duration, due to the bottlenecking of phonons on resonance with the electronic transition [9]. The amplitude of the R_2 luminescence, normalized to the R_1 intensity, is 0.7×10^{-4} , which is about 10 times smaller than the ratio measured along the a -axis [3]. The transverse profile of the additional contribution in the LA-luminescence is shown in Fig. 1(c), showing a Gaussian profile with a waist of about 0.4 mm. This corresponds favorably to the width of the pump laser beam at the position of the transducer, indicating a divergence angle of less than a degree. This directionality, together with

the highly nonlinear dependence of this signal on pump fluence (see Ref. [3]), proves that the additional luminescence is induced by the strain solitons.

The soliton-induced R_2 luminescence is the strongest when the excited zone is positioned just on the inside edge of the crystal, but partially overlapping with this edge (cf. Fig. 1(a)). We measured precisely the decay of this signal into the crystal as well as beyond the edge of the crystal. The results, presented in Fig. 2, show a strong decay of the luminescence into the crystal within $\sim 200 \mu\text{m}$ from the crystal surface, followed by a more slowly decreasing tail up to $\sim 1 \text{ mm}$. In the other direction, the intensity decreases much more rapidly due to the reduction of the overlap between the probe volume and the crystal.

For the explanation of the enhancement of the soliton-induced luminescence near the surface we consider the known theories of mode conversion of phonons reflecting at a rough interface. Much work has been done on this problem, related to the enhanced transmission of phonons through an interface, known as Kapitza resistance. Following the work of Nakayama [11], we consider the two

processes of diffusive scattering into either bulk or surface modes. The latter have a mode-conversion time back into bulk waves of $\tau_R = 100\nu^{-5}$, with ν the phonon frequency in GHz, which for 0.87 THz phonons corresponds to a lifetime of 0.2 ps. We consider the endface as a Lambertian surface, diffusively reflecting the phonon back into the crystal. We simulate the strong anisotropy in the coupling strength between strain and the electronic transitions by a dipolar form $f(\theta, \phi) = |\sin \theta|$, with θ the angle with the c -axis. Combined with the $1/r^2$ dependence of a hemispherical radiation source, this gives an interaction density for a point source as depicted in Fig. 2(inset), with approximately circular symmetry around the c -axis. For a cylindrical detection zone perpendicular to this axis, we may integrate the source function over the cartesian x -direction, and after convolution with the Gaussian (y, z) beam-profile (dashed line in Fig. 2) we arrive at the luminescence intensity, assuming an identical bottlenecking factor at all positions in the crystal. Excellent agreement is obtained with our experimental data using this model (line in Fig. 2).

The interaction mechanism resulting in the additional R_2 luminescence near the endface cannot be the same as that in Ref. [3], as the solitons are pulled apart by dispersion within several micrometers after the reflection from a free surface. Additionally, surface roughness results in a distorted wavefront on the nanometer scale, which is of the order of the soliton width. These disturbances together induce a severe destruction of the coherence of the strain pulse directly after reflection. The sudden decrease of the peak strain amplitude in this process disables the nonlinear coupling of the frequency components, ‘freezing in’ the acoustic spectrum as it was just before the reflection. Thus, the interaction with the electronic states after the diffuse reflection of the strain packet should be considered like that of an incoherent phonon spectrum, exhibiting accordingly different dependence on concentration for two-level systems than that proposed for ultra-short solitons in Ref. [3]. A more detailed analysis of the characteristics of these dependences at this point, however, is prevented due to the low luminescence yields in the current experiment.

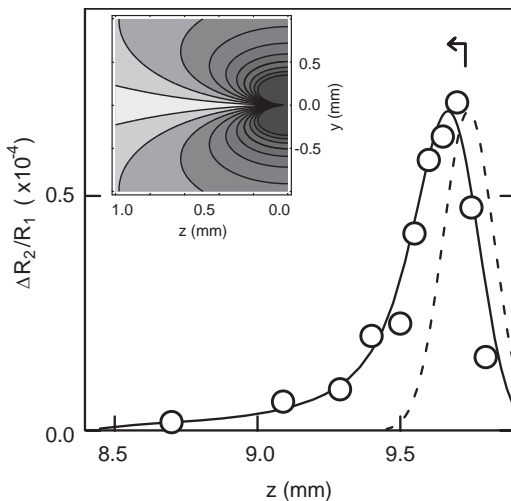


Fig. 2. Soliton-induced R_2 intensity normalized to R_1 as a function of distance from the transducer, (○) experimental data, arrow denotes exact surface position; model calculations for localized surface excitation (dash) and diffusive scattering (line). Inset: Electron–phonon interaction density in the crystal for point source at $(0,0)$, as used in a diffusive scattering model.

3. Conclusion

In conclusion, we have studied the interaction of ultrashort strain solitons and the electronic $\bar{E}(^2E) - 2\bar{A}(^2E)$ -transition along the ruby c -axis. The absence of soliton-induced luminescence in the bulk of the crystal is consistent with the suppression of the interaction due to the selection rules of the trigonal point group. The observation of a signal near the far end of the crystal has been explained by the increased coupling of the reflected wave due to mode conversion at the surface roughness. The decay of this luminescence intensity into the crystal can be explained by a hemispherical distribution of scattered phonons in combination with a dipolar form for the anisotropy of the transition matrix element.

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