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Comment on "Observation of anomalous acoustic phonon dispersion in $SrTiO_3$ by broadband stimulated Brillouin scattering" [Appl. Phys. Lett. 98, 211907 (2011)]

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In Ref. 1, the authors apply the picosecond acoustic method to the measurement of longitudinal acoustic phonons in (001) strontium titanate (STO) single crystals. They use a spectroscopic setup recently introduced by some of them and which enables the simultaneous probing of the sample reflectivity over a wavelength range. The probe laser detects the phonon propagation as an oscillation in the time-domain known as a Brillouin oscillation.[2] It is the result of interferences between the probe that is reflected from the interfaces of the sample and the light partially reflected from the strain pulse. At normal incidence, the period can be written as $T = \lambda/2nv$, where λ is the probe wavelength, n is the optical index, and v is the sound velocity. By measuring the oscillation period over a wavelength range, the authors of Ref. 1 deduced the longitudinal sound velocity at various acoustic frequencies. Surprisingly, they found that the sound velocity is not constant but varies by 5% in the investigated phonon wave vector range.

The intent of this Comment is to warn readers that the conclusion is probably wrong due to the setup itself. We obtained new data which are very close to what is expected from a substrate, and no deviation of the sound velocity is observed. We performed experiments on a $10 \times 10 \times 0.5$ mm^3 bulk commercial (100) STO substrate[3] As in Ref. 1, the substrate was covered by a thin Al film in order to convert light pulses into acoustic pulses. We used a conventional pump and probe setup associated with a femtosecond laser tunable between 690 and 1040 nm at a repetition rate of 80 MHz with 120 fs optical pulse width. The probe is frequency doubled to reach the blue-UV range. Our data are obtained by tuning the laser wavelength by wavelength. The signal to noise ratio is better than using a spectroscopic setup which permits to study the response of the substrate over a wider wavelength range.

In Fig. 1, we plot our experimental data, the data from Ref. 1 and the theoretical data obtained from the Brillouin formula, sound velocity, and optical index are extracted from literature.[4,5] All data are superimposed in the visible range. On the contrary, in the UV range (below 450 nm), a discrepancy clearly appears: our data are still close to the theoretical line, whereas data from Ref. 1 present an unexpected behavior. We detect no change in sound velocity up to 120 GHz, a frequency much higher than that studied in Ref. 1. From that we conclude that the extraction of sound velocity in Ref. 1 is wrong. A Brillouin oscillation period is converted in sound velocity from wavelength and optical index. An error concerning the optical index cannot explain the observed effect since dispersion leads to a worse agreement between experimental data and theory. Error could come from the probe wavelength. The probe used in Ref. 1 has a wide spectrum and the wavelength is not as well known as in our setup. The authors mentioned in a note that they used some interferometric filters for the spectrometer calibration. We suspect that they used visible filters which could explain the agreement between all the data in such a range but did not use any filter in the UV range.

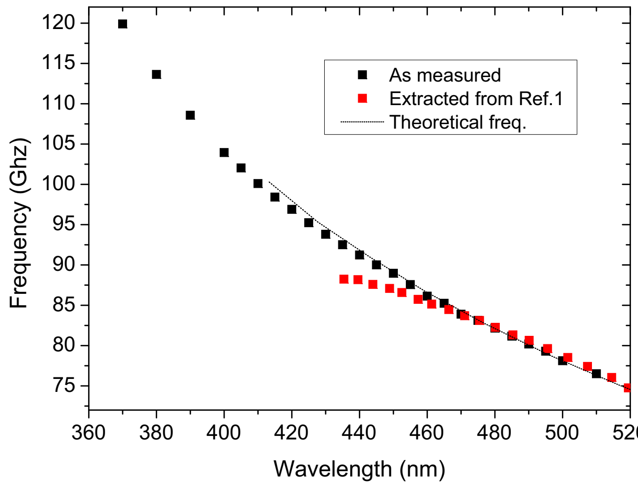

FIG. 1. Comparison between experimental and theoretical data.

To conclude, we think that there is no special acoustic dispersion effect in STO. Detection in ultrafast acoustics has been shown to be very sensitive to laser-wavelength.[6] A spectroscopic setup is a promising tool to take advantage of such effects but a special attention must be paid to wavelength calibration.

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