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Comment on “Visualizing coherent phonon propagation in the 100 GHz range: A broadband picosecond acoustic approach” [Appl. Phys. Lett. 98, 011901 (2011)]

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Reference 1 presents a new implementation of a previous picosecond optical technique (POT).^{2,3} It allows addressing a range of sound frequencies ν in single acquisition sweeps. The authors measured the sound-attenuation linewidth Γ of fused silica and concluded that it exhibits an acoustic crossover at $\nu \approx 170$ GHz. Although the role of multiple interferences⁴ is mentioned, these are not included in the analysis. To justify this neglect, the authors show that the squared linear-correlation coefficient r^2 of their least-square fits is nearly one around the purported crossover. The main intent of this Comment is to warn readers that accounting for multiple interferences is *essential* in POT if one desires better than just an order-of-magnitude estimate of the sound-attenuation coefficient, even if r^2 seems close to one. A second objective is to briefly discuss the acoustic attenuation of fused silica, pointing out the absence of POT evidence for any new crossover at frequencies up to 300 GHz.

To illustrate the effect of multiple interferences, we calculate, for an initially smooth linewidth function $\Gamma(\nu)$, the result of the simplified evaluation procedure in the experimental conditions of Ref. 1. Our choice for $\Gamma(\nu)$ is to adjust the approximative Γ of Ref. 1 to the anharmonic law $\Gamma(\nu) \propto \nu^2$. Indeed, at room temperature and at these frequencies anharmonicity should be the leading damping mechanism.⁵ This gives the dashed line in Fig. 1. We calculate then the ratios $A_{\text{Si}}/A_{\text{SiO}_2}$ defined in Ref. 1 for their six sample thicknesses, taking full account of interferences,⁴ and using the refractive index values of titanium.⁶ We obtain thereby a series of diagrams similar to Fig. 3(b) of Ref. 1, except that there are no random errors on the data points. The crucial approximation is the fitting of these diagrams to straight lines, as done in Ref. 1, which amounts to neglecting multiple interferences. This leads to the calculated apparent values Γ_{cal} and the related squared correlation coefficient r_{cal}^2 shown in Fig. 1. One first notes the rather large excursions of Γ_{cal} away from $\Gamma(\nu)$. The exact position of the wiggles strongly depends on the precise sample thicknesses. The behavior is remarkably similar to the Γ reported in Ref. 1. The “crossover” around 170 GHz, absent from the initial $\Gamma(\nu)$, can be fully explained by the neglected interferences. Inter-

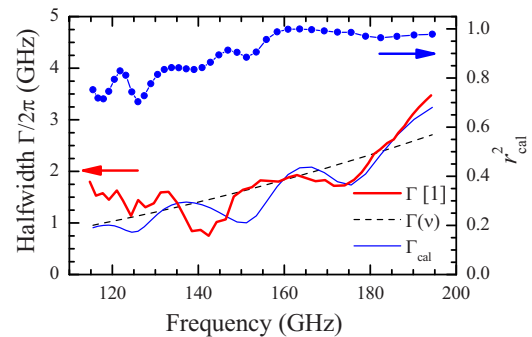


FIG. 1. (Color online) The measured Γ from Ref. 1, the fitted $\Gamma(\nu) \propto \nu^2$, and the calculated Γ_{cal} and r_{cal}^2 (see text).

estingly, the oscillatory features in r_{cal}^2 , while unrelated to random errors, also closely resemble the r^2 in Ref. 1. Obviously r^2 is not a good criterion to validate the simplified evaluation procedure.

The true acoustic damping of the silica films measured in Ref. 1 is probably closer to the dashed line in Fig. 1 than to the reported red line, and it shows no convincing crossover. This dashed line can be compared to recent POT results for which all interferences were taken into account.⁷ The ν dependence of Γ is there fully explained by thermally activated relaxations plus anharmonicity, extrapolating the low frequency regime and without any new crossover up to 320 GHz.^{5,7} It is interesting that the latter half widths fall somewhat below the dashed line. This may relate to different preparations of the oxide layers. Also, with the broadband technique,¹ the enhanced acquisition rate must be at the cost of a higher transient heating of the samples that can affect the results.

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