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Optimized determination of elastic constants of crystals and their uncertainties from surface Brillouin scattering

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ABSTRACT

Surface Brillouin scattering of light allows the angular-dependent velocities of Rayleigh surface acoustic waves (SAW), pseudo-SAW and longitudinal lateral waves (L) on the surface of an opaque crystal to be measured, and the elastic constants thereby determined. Closed form expressions exist for the surface wave velocities in high symmetry directions on crystallographic symmetry planes, and these have been exploited in the past for obtaining the values of the elastic constants. This paper describes a procedure for obtaining an optimized set of elastic constants from SAW, pseudo-SAW and L velocities measured in arbitrary directions in the (001) and (110) surfaces of cubic crystals. It does so by affecting a linearization of the numerically determined angular-dependent SAW and pseudo-SAW velocities near the best fit, and using analytic expressions for the L velocity. The method also generates covariance ellipsoids, from which the uncertainties in the determined values of the elastic constants can be read off. The method is illustrated using surface Brillouin scattering data to obtain the room-temperature elastic constants C_{11}, C_{12} and C_{44} of the cubic crystals $VC_{0.75}$ and Rh_3Nb.

1. Introduction

Surface Brillouin scattering (SBS) is widely used for the determination of the near-surface elastic constants of opaque crystals and thin supported films [1,2]. The commonly employed back scattering geometry is illustrated in Fig. 1. Laser light of wave vector k_i is incident on the surface of a highly polished surface of an opaque specimen. A small fraction of the back-scattered light is observed to have undergone a slight frequency shift $\omega = \pm v k_{//}$ due to thermodynamically fluctuating surface corrugations of velocity v and $k_{//} = 2k_i \sin\theta$. Scattering from highly opaque samples is dominated by the surface ripple mechanism, for which at room temperature and above the scattering cross section is given by [1,2]

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{AT}{\omega}\,\mathrm{Im}\,G_{33}(k_{\parallel},\omega). \qquad (1)$$

Here $G_{33}(\mathbf{k}_{\parallel},\omega)$ is the elastodynamic Green's function for the medium pertaining to force and displacement response normal to the surface, T is the absolute temperature and A is a constant depending on the scattering geometry and incident light. Fig. 2 shows an example of a calculated $G_{33}(\mathbf{k}_{\parallel},\omega)$. The method of calculation is described in Ref. [3].

SBS spectra are usually dominated by a sharp peak arising from inelastic scattering by Rayleigh surface acoustic waves (SAW) or pseudo-SAW. In any direction in the surface, Rayleigh SAW are subsonic with regard to all bulk waves in that direction and are a superposition of evanescent partial wave components that satisfies the free surface boundary conditions. In principal it is an infinitely sharp resonance. A small amount of damping is imposed in the calculation of G_{33} [3] to broaden the delta function imaginary part at the Rayleigh pole and render it visible in Fig. 2 and elsewhere in the paper. Pseudo-SAW are supersonic, being located in the bulk wave continuum, and have a small partial bulk wave component into which they radiate, causing their attenuation and finite spectral broadening. In this paper we restrict our attention to the type of pseudo-SAW, which for certain specific directions in the considered surface, uncouples from the bulk wave continuum and becomes an un-attenuated supersonic SAW. Near to such directions pseudo-SAW are narrow resonances [4,5] suited for use in elastic constant determinations. There are reports of pseudo-SAW on the (110) surfaces of crystals such as GaAs which do not share this feature [6,7].

In principle, the angular dependence of the SAW velocity, measured with sufficient accuracy on one or more surface crystallographic orientations, provides sufficient information to infer most or all of the elastic constants of a cubic and other high symmetry crystal. However, the sensitivity of SAW velocities to "longitudinal

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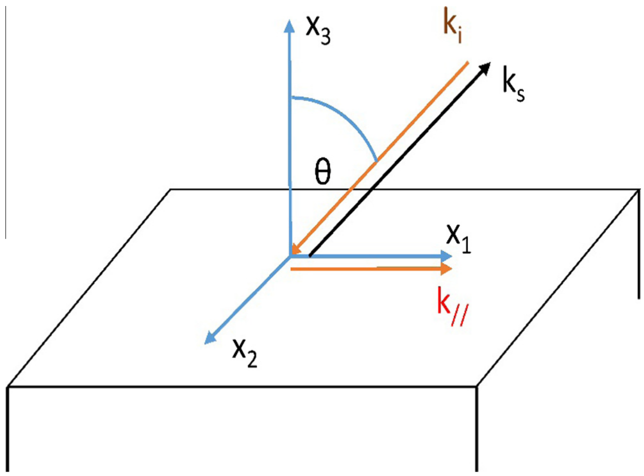

Fig. 1. Back-scattering geometry in surface Brillouin scattering.

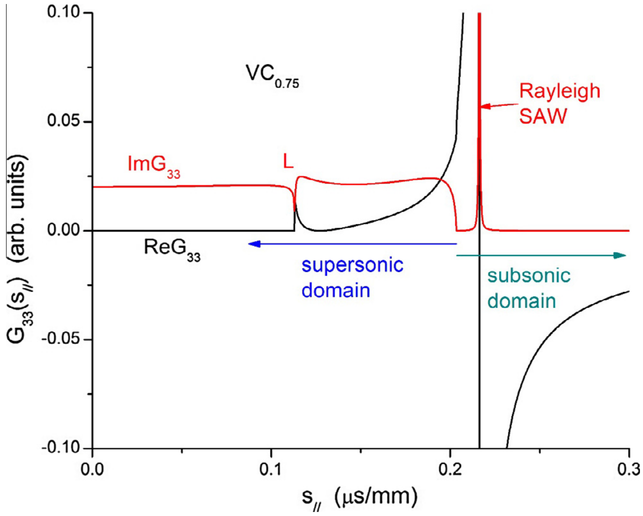

Fig. 2. G_{33} represented as a function of slowness $s_{\parallel} = k_{\parallel}/\omega$ with frequency ω fixed, for the [100] direction in the $VC_{0.75}$ (001) surface.

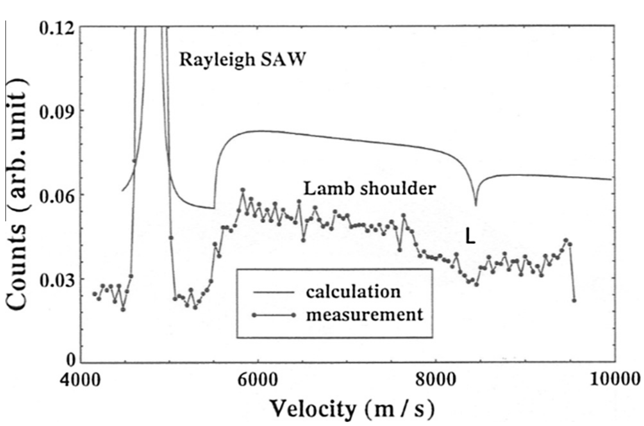

Fig. 3. SBS spectrum for the $VC_{0.75}$ for the $[1\bar{1}0]$ direction in the (110) surface. From Ref. [8].

wave" elastic constants such as C_{11} and $C_{12} + 2C_{44}$ is limited, and where possible SAW data is supplemented by measurements of the longitudinal bulk wave threshold (L), a characteristic dip also known as the "longitudinal lateral wave", in the continuous portion of the SBS spectrum, the Lamb shoulder, see Fig. 3. For practical reasons much larger data collection times are required to locate the L threshold as compared to the SAW peak. Commonly therefore the data sets from which the elastic constants are inferred consist of large numbers of SAW velocity measurements for different configurations and only a few or even a single L velocity measurement.

In this paper we set out a strategy for the optimized determination of the three elastic constants C_{11}, C_{12} and C_{44} of cubic crystals and their experimental uncertainties for this scenario. We illustrate our approach by determining the elastic constants and their uncertainties for two crystals $VC_{0.75}$ and Rh_3Nb, using data we have previously published in Refs. [8,9]. Prior knowledge gained on these two materials has provided starting values of the elastic constants for the optimization procedure. The location of these two materials in a plot of C_{11}/C_{44} vs. C_{12}/C_{44} is shown in Fig. 4. This plot distinguishes the classes of materials displaying pseudo-SAW in either the (001) or (110) surfaces.

The Zener anisotropy factor [10] for $VC_{0.75}$ is $\eta = 2C_{44}/(C_{11} - C_{12}) < 1$, and this crystal falls in a class of materials for which the Rayleigh SAW is to be observed in all directions in both the (001) and (110) surfaces, and there is no pseudo-SAW at all. This is illustrated by Fig. 5a, which is a gray scale representation of Im $G_{33}(s_{\parallel})$ for the (001) surface of $VC_{0.75}$, which reveals a distinct Rayleigh SAW in all directions and no pseudo-SAW. Measured data from Ref. [8], comprising SAW measurements in a number of directions in both these surfaces and a single measurement of the L velocity in the (110) surface is used for the optimized determination of the elastic constants of $VC_{0.75}$.

The Zener anisotropy factor for Rh_3Nb is $\eta > 1$, and this crystal falls in a class of materials for which in the (001) plane the Rayleigh SAW is to be clearly observed on either side of the [100] direction, and fading in intensity to zero towards the [110] direction, while within an angular range on either side of the [110] direction a pseudo-SAW dominates the scattering, see Fig. 5b. There is no measured data available for the (110) surface of this crystal. The (100) SAW measurements, supplemented by 5 measured L velocities in directions on either side of [100], provide the data for the optimized determination of the elastic constants of Rh_3Nb.

We determine the elastic constants and their uncertainties by a least squares fit, minimizing

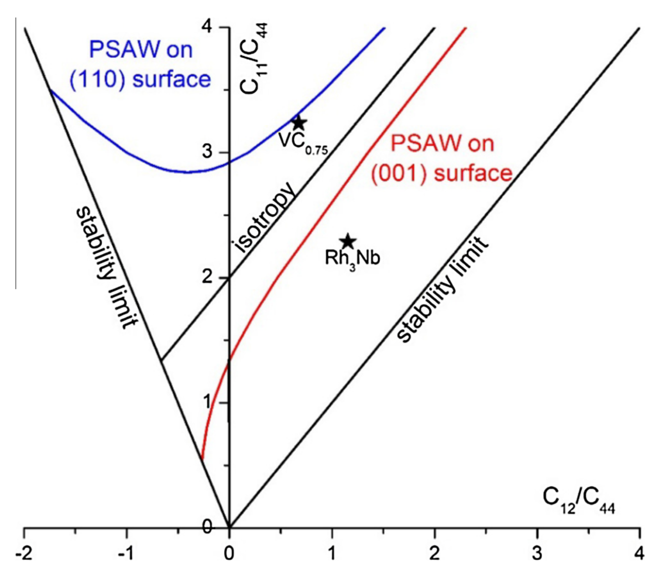

Fig. 4. Existence domains for pseudo-SAW interspersed with supersonic SAW on the (001) or (110) symmetry plane surfaces of cubic crystals.