

TEMPERATURE DEPENDENCE OF THE HYPERSONIC VELOCITY IN A SINGLE CRYSTAL OF ARGON*

H. MEIXNER, P. LEIDERER and E. LÜSCHER

Physik-Department der Technischen Universität München, Munich, Germany

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The temperature dependence of the longitudinal sound velocity in the [100]-direction has been measured in a single crystal of argon in a temperature range from 4.2°K to 77°K by stimulated Brillouin scattering. Using the sound velocity, together with previous data for the bulk modulus and the Debye temperature, the elastic constants C_{11} , C_{12} and C_{44} was determined.

The noble gas crystals are of interest as model crystals for lattice dynamics. One can obtain valuable information on lattice dynamics through the knowledge of sound velocity. Gsänger et al. [1, 2] and Keeler et al. [3] have reported measurements of the sound velocity using a standard pulse-echo technique on single-crystals of argon.

In this letter we present the first measurements of the temperature dependence of the longitudinal sound velocity in a single crystal of argon by stimulated Brillouin scattering. The temperature dependence was determined in the range from 4.2°K to 77°K.

In our experiment we used an optical arrangement as described in [4, 5, 7] to observe stimulated Brillouin scattering in solid argon. The beam of a single mode giant pulse ruby laser (peak power 5 MW, half width 20 nsec, beam divergence about 1 mrad, $\lambda = 6943 \text{ \AA}$) was focused by a lens into the argon sample. The frequency shift $\Delta\nu$ (Hz) of the backscattered stimulated Brillouin light was detected by a Fabry-Perot interferometer within an accuracy of about $\pm 0.3\%$. $\Delta\nu$ is related to the hypersonic velocity V by $\Delta\nu = 2nV/\lambda$. The value of the refractive index n was taken from ref. [8, 9].

The argon sample (fcc structure) was grown from a natural isotopic mixture at an impurity concentration of less than 4 ppm. The growing technique is described by Gsänger et al. [10].

The crystal was examined by Bragg scattering of neutrons at the research reactor in Munich. Neutron scattering experiments and neutron photography at 4°K showed the presence

of a single crystal, with an estimated volume of about 11 cm^3 . The single crystal of argon had a diameter of approximately 22 mm and a length of 31 mm. It was possible to show within an error of $\pm 1^\circ$ that Brillouin scattering occurred in the [100]-direction of the single crystal.

Our results are plotted in fig. 1. All data were taken by shooting into the same spot of the sample without laser-induced damage. The values of the

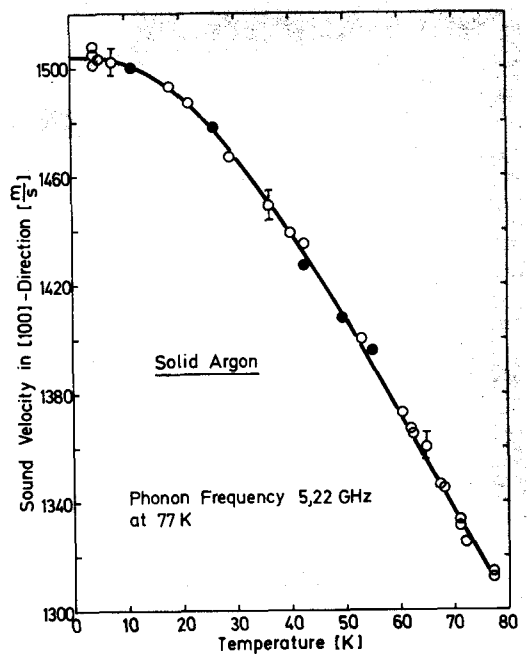


Fig. 1. Sound velocity in single-crystal argon versus temperature. The longitudinal sound wave propagating in the [100]-direction was studied. The open and full circles refer to measurements at decreasing and increasing temperature, respectively.

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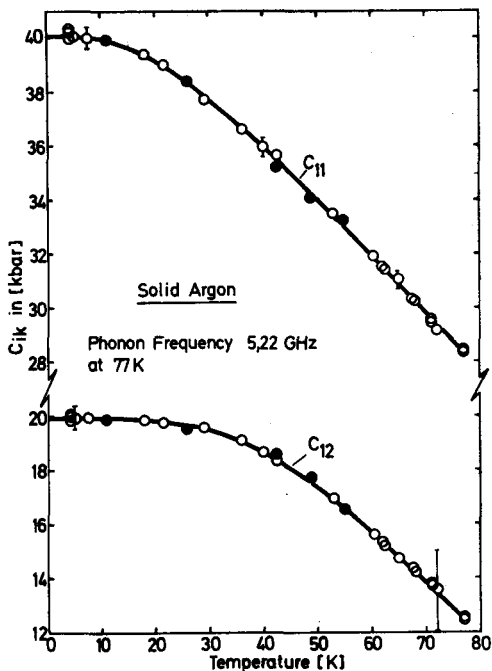


Fig. 2. Elastic constant C_{11} in a single-crystal argon versus temperature. The open and full circles refer to measurements at decreasing and increasing temperature. The accuracy of the determination for C_{11} is 1.0 %. We have also plotted C_{12} against the temperature as determined from eq. (2).

elastic constant C_{11} shown in fig. 2 were obtained from these data by using the formula

$$V_{\text{long}}^2 [100] = C_{11} \quad (1)$$

using the relation

$$B^{\text{ad}} = \frac{1}{3}(C_{11} + 2C_{12}) = 1/(\chi^{\text{is}} - \beta^2 VT/C_p) \quad (2)$$

we could determine the elastic constant C_{12} , where χ^{is} is the isothermal compressibility; β the thermal expansion coefficient, V the molar volume, T the temperature and C_p the isobaric specific heat c_p . All experimental data are taken from ref. [11]. In fig. 2 we have also plotted C_{12} versus the temperature. From the results for C_{11} and C_{12} we calculated C_{44} at $T = 0$ K, using a relation given by De Launay [12]

$$\Theta_0^{\text{D}^3} = \left(\frac{h}{K}\right)^3 \frac{9}{4\pi V_{\text{EZ}}} \left(\frac{C_{44}}{\rho^{3/2}}\right)^{3/2} \frac{9}{18+\sqrt{3}} f(S, t) \quad (3)$$

where

$$S = \frac{C_{11} - C_{44}}{C_{12} + C_{44}} \text{ and } t = \frac{C_{12} - C_{44}}{C_{44}},$$

h/k is the ratio of Planck's constant to Boltzmann's constant, V_{EZ} is the volume of the unit cell, ρ the density and Θ_0^{D} , the Debye temperature for $T = 0$ K. The factor $f(S, t)$ is a function of the anisotropy of the crystal and has been tabulated [12]. Using eq. (3) we obtain

$$C_{44} = (22.8 \pm 1.8) \text{ kbar,}$$

for $T = 0$ K.

From our calculation for the elastic constant we obtain for the elastic anisotropy A at $T = 0$ K

$$A = 2C_{44}/(C_{11} - C_{12}) = 2.28 \pm 0.29,$$

and for the deviation δ from the Cauchy relation

$$\delta = (C_{44} - C_{12})/C_{12} = 0.14 \pm 0.11.$$

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