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The following is the complete title, byline, and abstract for the Letter by Lubzens, Shanabarger, and Schultz; see Notice, p. 1717, this issue. The original appeared in *Phys. Rev. Lett.* **29**, 1387 (1972).

Observation of a Frequency Dependence in the Conduction-Electron Spin Resonance of Al, Cu, and Ag, Interpreted as a New Many-Body Effect in Metals with g Anisotropy*

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The linewidth and g value for the conduction-electron spin-resonance-transmission line shape of aluminum was observed to be frequency and temperature dependent. The data are interpreted as a consequence of motional narrowing and many-body exchange in metals with g anisotropy. The data analysis yields a measure of the g anisotropy, a many-body parameter for the spin part of the Landau correlation function, and the behavior of a relevant relaxation time.

In the last line of the caption of Fig. 3, the reference should be Ref. 9 instead of Ref. 12.

The following Letter is reprinted in full; see Notice, p. 1717, this issue. The original appeared in *Phys. Rev. Lett.* **29**, 1454 (1972).

**Elastic Constants of Argon and Neon by Brillouin Scattering
 from Single Crystals near Their Triple Points***

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The adiabatic elastic constants of single crystals of neon at 24.3 K and of argon at 82.0 K have been determined by Brillouin scattering. For argon the values are $C_{11}=2.33 \pm 0.08$, $C_{12}=1.49 \pm 0.06$, $C_{44}=1.17 \pm 0.07$, in units 10^{10} dyne/cm², with the elastic anisotropy $A=2.80 \pm 0.60$; for neon, the values are $C_{11}=1.175 \pm 0.020$, $C_{12}=0.740 \pm 0.020$, $C_{44}=0.595 \pm 0.015$, and $A=2.74 \pm 0.25$.

The elastic constants of the rare-gas solids, and their dependence on temperature, provide sensitive tests of recent theories of lattice dynamics.¹⁻³ Especially important are values of the constants at high temperatures in order to check the anharmonicities of assumed interatomic potentials^{4,5} and the possible relevance of

many-body forces.⁶ Neon, because of its small atomic mass and consequent large-amplitude lattice vibrations, requires more detailed calculations at high temperatures, and therefore should provide a very stringent test of theory. However, only one determination of its elastic constants has been reported to date,⁷ based on

neutron scattering measurements for a crystal at 4.7 K and for a second crystal under pressure at 4.7 and 25 K. Argon is the most thoroughly studied of the rare-gas solids both experimentally and theoretically. In spite of this activity with argon, significant difficulties remain with our present knowledge of its elastic constants.⁸⁻¹³ The experimental values do not show good agreement among themselves nor is there satisfactory agreement between experimental and theoretical values.^{1,2}

We report here on the determination of the adiabatic elastic constants of neon and argon near their triple points, based on Brillouin scattering experiments with single crystals. The experimental techniques were essentially the same as those described by Gornall and Stoicheff¹⁴ in their determination of the elastic constants of Xe single crystals. Crystal samples were grown from the liquid in equilibrium with the vapor, in cylindrical cells (~2 mm i.d. and 10 mm long) mounted with their axes vertically in the tail sections of suitable Dewars. Laue x-ray transmission photographs were used to check that the samples were single crystals, and to establish (within ~30') their orientation as specified by the Euler angles θ, φ, χ (with φ corresponding to rotation about the vertical axis). Eight single crystals of Ne grown at 24.3 K and one single crystal of Ar at 82.0 K were used in the present study.

For the experiments with solid argon, radiation from a stabilized, single-frequency, He-Ne laser, operating at 6328 Å with 6 mW output, was directed along the vertical axis of the cell and focused in the crystal. Light scattered at 90° was analyzed with a pressure-scanned Fabry-Perot interferometer. Solid neon, because of its smaller polarizability, is a much less efficient light scatterer than solid argon, by a factor of ~20. Thus, improvements in laser excitation and in light detection were necessary: A stabilized, single-frequency, Ar⁺ laser emitting ~50 mW at 4880 Å was used, along with a piezoelectrically scanned interferometer and photon-counting detection system coupled to a multichannel analyzer for storage of the data during several thousand sweeps of the spectrum.

Brillouin spectra were recorded for all of the crystals at several different orientations about their vertical axes. For one of the Ne crystals, spectra were obtained at eleven different rotation angles φ , and for the Ar crystal at eight angles φ . The observed Brillouin spectra contained the longitudinal component and generally

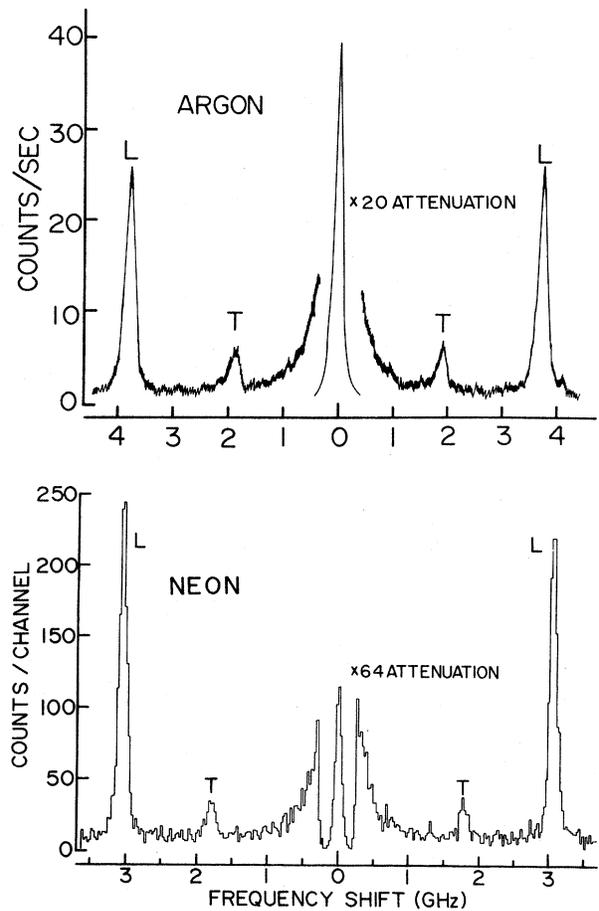


FIG. 1. Typical Brillouin spectra of Ar and Ne single crystals.

one transverse component, as shown in the spectra of Fig. 1. For a cubic crystal, of course, two transverse components are expected in addition to the longitudinal component. However, it can be shown from the relative values of the photoelastic constants of Xe,¹⁴ which are also representative of those of Ne, Ar, and Kr, that the "fast" transverse component is of extremely low intensity except for a very small range of orientations. None of these orientations was obtained in the present experiments.

The measured values of the frequency shifts for the eleven orientations of the Ne crystal mentioned above, and eight orientations of the Ar crystal, are shown in Fig. 2. Each of the frequency shifts was analyzed according to the well-known Brillouin equation

$$\Delta\nu_i = \pm 2\nu_0(V_i/c)n \sin(\theta/2)$$

to determine the velocities V_i of the thermal waves. The frequency shifts $\Delta\nu_i$ were measured

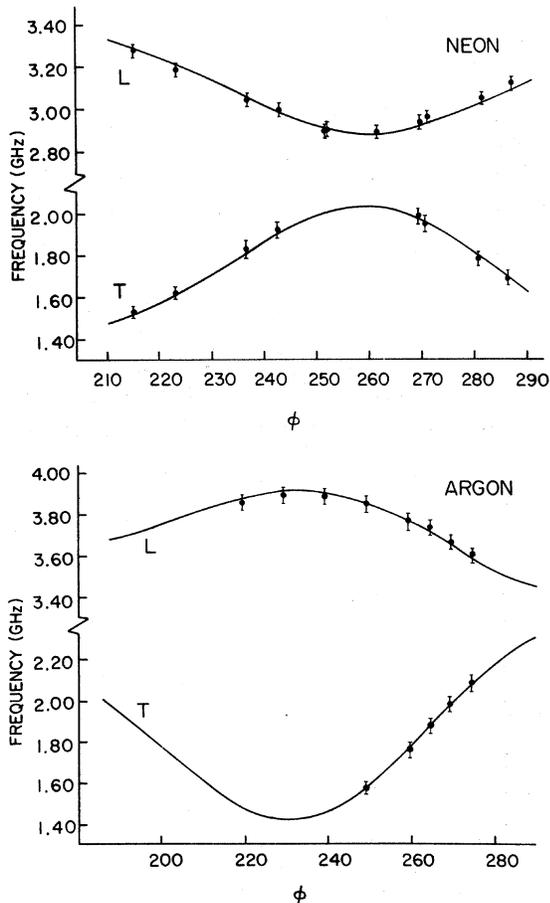


FIG. 2. Observed and calculated frequency shifts versus rotation angle ϕ . The indicated errors on the experimental points include inaccuracies in measurements of orientation angles as well as of frequency shifts. The curves are calculated values based on the elastic constants given in Table I.

to an accuracy of 0.5% for longitudinal and 1% for transverse components; the incident frequencies ν_0 of $15\,798\text{ cm}^{-1}$ at 6328.2 \AA and $20\,492\text{ cm}^{-1}$ at 4879.9 \AA are accurately known; and the scattering angles θ were accurate to $\pm 15'$. For Ar, a value of the refractive index $n(6328\text{ \AA}) = 1.2674$ was determined from the measurements of Sinnock and Smith.¹⁵ A measured value for the refractive index of solid Ne is not available. How-

ever, a calculation based on the Clausius-Mossotti relation, and on the measurements of the static dielectric constant of liquid Ne near the triple point by Bewilogua, Handstein, and Hoeger¹⁶ gives $n = 1.107 \pm 0.003$. For calculation of the elastic constants, the following values of the densities were used: $\rho(\text{Ne}) = 1.4371 \pm 0.0015\text{ g/cm}^3$,¹⁷ and $\rho(\text{Ar}) = 1.627 \pm 0.001\text{ g/cm}^3$.¹⁸

A least-squares analysis of all of the data was carried out yielding the values of the adiabatic elastic constants given in Table I. Included in Table I are values of the elastic anisotropy parameter $A = 2C_{44}/(C_{11} - C_{12})$, the adiabatic bulk modulus $B_s = \frac{1}{3}(C_{11} + 2C_{12})$, and the Grüneisen parameter γ . Larger errors are quoted for the constants of Ar than for those of Ne since measurements were made on only one crystal of Ar compared with eight of Ne. An indication of the overall accuracy of the elastic constants listed in Table I is given by the close fit of the calculated frequency shifts to the experimental measurements in Fig. 2.

The only experimental values of all three elastic constants of neon available for comparison are the "zero-sound" values reported by Leake *et al.*⁷ at 4.7 K. Their results are $C_{11} = 1.69$, $C_{12} = 0.97$, and $C_{44} = 1.00$, all in units of 10^{10} dyne/cm^2 , giving $A = 2.8$. The elastic constants have thus been shown to decrease by 25 to 40% in the temperature range 4.7 to 24.3 K, emphasizing the importance of anharmonicity in solid neon. Other experimental measurements for comparison include the longitudinal and transverse sound velocities in polycrystalline neon by Bezuglyi, Plakhotin, and Tarasenko,¹⁹ and by Balzer, Kupperman, and Simmons.²⁰ The latter authors deduced a value of $B_s = 0.758 \times 10^{10}\text{ dyn/cm}^2$, which does not agree with our value within their quoted error limit of 5%. Finally, for neon, the value $\gamma = 2.65$ indicates an approximately constant value in the temperature range 4 to 24.3 K rather than a rapid decrease above 16 K as previously reported.^{17,19}

The results of three different investigations based on ultrasonic measurements on argon sin-

TABLE I. Adiabatic elastic constants of Ne and Ar.

	C_{11} (10^{10} dyn/cm^2)	C_{12} (10^{10} dyn/cm^2)	C_{44} (10^{10} dyn/cm^2)	A	B_s (10^{10} dyn/cm^2)	γ
Ne (24.3 K)	1.175 ± 0.020	0.740 ± 0.020	0.595 ± 0.015	2.74 ± 0.25	0.885 ± 0.020	2.65 ± 0.13
Ar (82.0 K)	2.33 ± 0.08	1.49 ± 0.06	1.17 ± 0.07	2.80 ± 0.60	1.77 ± 0.07	2.68 ± 0.13

gle crystals near the triple point are available for comparison with the values of the elastic constants presented in Table I. The most complete and extensive measurements are those reported recently by Keeler and Batchelder.¹⁰ Their data yield the values $C_{11} = 2.702$, $C_{12} = 1.390$, and $C_{44} = 0.887$ (10^{10} dyn/cm²) at 82 K, which differ from the present determinations by ~10%. Furthermore, they obtained a value of $A = 1.35$ compared with the present value 2.80, while the two earlier experiments^{3,9} yielded values of $A < 1$ opposite in sense to theoretical predictions. A recent experiment on stimulated Brillouin scattering¹³ from argon in the $\langle 100 \rangle$ direction yielded the constant $C_{11} = 2.83 \times 10^{10}$ dyn/cm², at 77 K. This is ~20% higher than the present value. There have also been two investigations^{11,12} on neutron scattering from argon single crystals. These were carried out at 4 K, and therefore the results are not directly comparable with the present values. However, when combined with the present values, they reveal decreases of ~25%, 10%, and 50% for C_{11} , C_{12} , and C_{44} , respectively, in the temperature range of 4 to 82 K, indicating considerable anharmonicity in solid argon.

A comparison of the present experimental values with available theoretical values leads to the result that theoretical calculations based on a Lennard-Jones 6-12 potential give surprisingly good agreement. Such a potential also gives good agreement between the experimental and theoretical values of the elastic constants of xenon^{2,14} near the triple point. In particular, calculations based on a 6-12 potential by Holt *et al.*¹ on Ne and Ar, and by Klein and Murphy² on Ar and Xe show good agreement with the experimental values for all three solids. In calculations by Klein and Murphy² on Ar, with the best available potential, the agreement is not as good: The calculated values of C_{11} and C_{12} differ from the experimental values by ~1.5 times the experimental error. These results are surprising since recent work on molecular-beam experiments with Ne₂²¹ and Ar₂,⁵ and on vibrational structure in electronic spectra of Ar₂,²² have shown that the Lennard-Jones 6-12 potential does not adequately describe²³ the pairwise forces in Ar₂ and Ne₂.

In summary, the present research has led to the determination of accurate values for the elastic constants of Ne and Ar near their triple points. The strength of the present method lies in the knowledge that measurements were carried out on single crystals. In this respect, these experiments differ from many of the earlier investiga-

tions, especially those based on ultrasonic measurements, which may explain the disagreement with earlier values of the elastic constants. Finally, this work has shown the desirability of further theoretical investigations of the elastic properties of the rare-gas solids, and in particular, of solid Ne.

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