

FIG. 1. Experimental spectra of light scattered from thermal surface vibrations on a liquid (methanol at 25°C) at three different surface wavelengths: (a)  $\Lambda = 92\lambda/n$ , (b)  $\Lambda = 77\lambda/n$ , and (c)  $\Lambda = 47\lambda/n$ , where  $\lambda = 4880 \text{ \AA}$  and  $n = 1.33$ . The arrows and brackets indicate the frequencies and linewidths calculated from classical theory of surface vibrations, with static values of surface tension and viscosity used. The instrumental bandwidth, including aperture broadening, is estimated to be 3.4 kHz, which is also indicated.

be seen, these values are in reasonably good agreement with the experimental values, and it appears that for this liquid (methanol at 25°C)

the classical dispersion relation and static values of  $\sigma$  and  $\mu$  can be applied in the surface wavelength region considered here. A detailed discussion of experimental results in methanol and other liquids will be presented elsewhere.

\*This work was supported principally by the U. S. Navy (Office of Naval Research) under Contract No. N00014-67-A-0204-0019.

<sup>1</sup>W. Brouwer and R. K. Pathria, *Phys. Rev.* **163**, 200 (1967).

<sup>2</sup>For wavelengths shorter than a critical wavelength  $\Lambda_c = 1.16\pi\mu^2/\sigma\rho$ , the surface vibrations are overdamped and exhibit nonoscillatory decay. For the liquid considered here,  $\Lambda > \Lambda_c$ , where  $\Lambda_c \approx 0.20\lambda/n$ , and only underdamped, oscillatory vibrations occur, according to the classical theory. For further details see R. H. Katyl and U. Ingard, *Phys. Rev. Letters* **19**, 64 (1967), and V. G. Levich, *Physicochemical Hydrodynamics*, translated by Scripta Technica, Inc. (Prentice-Hall, Inc., Englewood Cliffs, N. J., 1962), p. 603.

<sup>3</sup>Katyl and Ingard, Ref. 2.

<sup>4</sup>C. H. Townes, in *Advances in Quantum Electronics*, edited by J. P. Singer (Columbia University Press, New York, 1961), pp. 3-11.

<sup>5</sup>J. B. Lastovka and G. B. Benedek, in *Physics of Quantum Electronics*, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966), p. 231; also see, *Phys. Rev. Letters* **17**, 1039 (1966).

### DIFFUSION OF EXCHANGE ENERGY IN SOLID $\text{He}^3$

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(Received 1 December 1967)

We report a method for determining the rate of diffusion of exchange energy in solid  $\text{He}^3$ . Over the temperature range 0.05–0.12°K, we obtain a temperature-independent diffusion constant  $D_E = (0.75 \pm 0.15)Ja^2$ , where  $a$  is the nearest-neighbor distance and  $J$  the exchange integral.

We report what we believe to be the first measurement of spatial diffusion of exchange energy in a paramagnet far above the magnetic transition temperature. The diffusion constant  $D_E$ , which is expected to be independent of temperature<sup>1</sup> and proportional to the exchange constant  $J$ , has been measured in the bcc phase of solid  $\text{He}^3$  using a new nmr technique. The experiment depended on the fact that the Zeeman system<sup>2</sup> (created by an external static magnetic field  $H_0$ ) was in good thermal contact with the exchange system.

The constant  $D_E$  is to be distinguished from the Zeeman diffusion constant  $D_Z$ , which in

$\text{He}^3$  was first determined by Reich<sup>3</sup> and has since been measured systematically by Thompson, Hunt, and Meyer.<sup>4</sup> Zeeman diffusion is a process wherein a temperature-independent transport of Zeeman energy results from the exchange mechanism and can be measured using well-known nmr techniques.<sup>5</sup> This differs from the present experiment since we determine the rate at which the total spin energy diffuses because of the exchange mechanism. The determination leads then to  $D_E$ .

The principle of the measurement is to heat the spin system locally and then watch this energy diffuse through the solid. In addition to

a static external field  $H_0$  in the  $z$  direction, there is applied a uniform gradient  $G = dH/dz$ . Thus the spatial dependence of the spin temperature can be monitored, since the Larmor frequency is spatially dependent. The exchange system can be heated in a thin slab of the solid via the closely coupled Zeeman system by applying rf power for several seconds. This "burns a hole" in the wide resonance line. If the Zeeman specific heat is chosen to be about one-fourth of the exchange specific heat, the effects of the Zeeman system are minimized while still allowing the exchange system to be heated. The spin system must be decoupled as well as possible from the lattice to prevent a loss of energy by relaxation. In Fig. 1 is shown the entire resonance line with a "hole" burned in the center. The wiggles in the center result from sweeping rapidly through the sharp "hole."

The presence of the wiggles makes this type of data exceedingly difficult to analyze, and suggests that pulsed nmr techniques would be more suitable. The same information is available with pulses, since the nuclear signal following a pulse is the Fourier transform of the line shape. The free induction signal will be composed of two parts: (1) a short-lived component due to the entire wide resonance line and (2) a long-lived component due to the narrow hole.

We assume that shortly after saturation the shape of the hole evolves into a Gaussian form. The time-dependent Gaussian, which is obtained by use of the Green's function for the one-dimensional diffusion equation, can be Fourier transformed. The resulting normalized function describes the free induction signal after

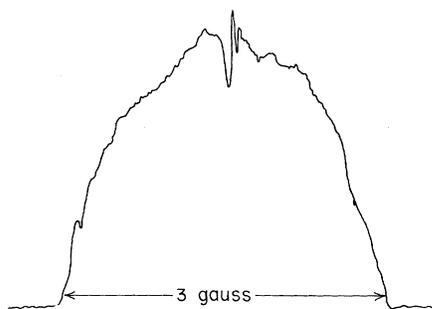


FIG. 1. The inhomogeneously broadened resonance showing the sharp hole with its associated wiggles. The width of the resonance corresponds to about 3 G for the gradient used (15.7 G/cm).

a pulse. It is found to be

$$G(t, \tau) = \exp\left[-\frac{1}{2}(\sigma_0^2 + 2\gamma^2 G^2 D \tau)t^2\right], \quad (1)$$

where  $\tau$  is the elapsed time since beginning the measurement (about 2-4 sec after saturation), where  $\sigma_0$  is the rms width at  $\tau=0$ , and where  $t$  starts at zero for each measuring pulse. Spin-lattice relaxation may be accounted for by multiplying the right-hand side by  $\exp(-\tau/T_1)$ . The combined diffusion constant  $D$  is obtained by analyzing the data according to Eq. (1).

The contribution of  $D_E$  to the measured  $D$  must be determined. Their relation is found by considering the two energy flow equations between the Zeeman and exchange systems and the lattice.<sup>1</sup> We define  $\tau_{ZE}$  and  $\tau_{EL}$  to be the Zeeman-exchange and exchange-lattice relaxation times respectively. Then, in the limit where  $(\tau_{ZE}/\tau) \rightarrow 0$  and  $(\tau_{EL}/\tau) \rightarrow \infty$ , the energy flow equations predict that the total spin energy diffuses with a constant

$$D = \frac{\omega_0^2 D_Z + 3J^2 D_E}{\omega_0^2 + 3J^2}. \quad (2)$$

Reflected in Eq. (2) is the fact that since the Zeeman and exchange systems are coupled, both contribute to the observed effect.

The experiment was performed at temperatures between 0.05 and 0.12°K in order to make  $\tau_{EL}$  sufficiently long ( $\lesssim 50$  sec)<sup>6,7</sup> to decouple the spins from the lattice. Measurements were performed at six exchange frequencies  $J/2\pi$  between 4 and 15 MHz. The latter were determined by measuring  $\tau_{ZE}$  and  $T_2$  and comparing these with an analysis of previous results.<sup>8</sup> Saturation with a 2-mG rf field for ~3 sec "burned a hole" in the nmr line. Then a series of small-angle ( $\sim 4^\circ$ ) pulses was used to observe the time-dependent width of the hole without heating the spin system significantly. In Fig. 2 is shown a partial series of Gaussian decays, where  $\tau=0$  for the top trace. The traces, each lasting 10 msec and occurring at 3-sec intervals, show beats which arise from mixing of the hole signal with a leakage signal from the rf generator (slightly detuned after saturation). The decreasing time constant due to the widening of the hole is clearly seen.

The data were well represented by Eq. (1). Errors are due largely to uncertainty in the gradient and to signal-to-noise problems because of the small pulses. Another systematic error arises because of the finite  $\tau_{ZE} \lesssim 0.2$

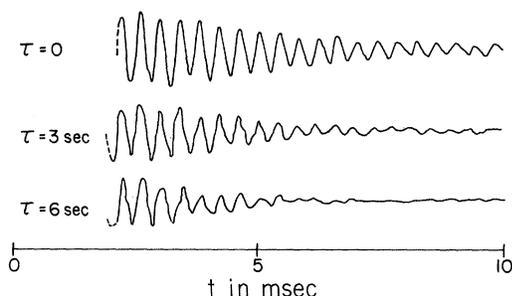


FIG. 2. The Gaussian free precession signals following  $4^\circ$  pulses every 3 sec. The top trace ( $\tau=0$ ) corresponds to  $\sigma_0=2.3 \times 10^2$  rad/sec. The decreasing time constant is apparent. For this sample,  $J/2\pi=14$  MHz.

sec. The Zeeman response will lag behind that of the exchange system by an amount depending on the rates of diffusion and the cross-relaxation time  $\tau_{ZE}$ . However, the time associated with diffusion is about an order of magnitude longer than  $\tau_{ZE}$ ; so the additional error is thought negligible. Once  $D$  is obtained using Eq. (1),  $D_E$  is determined by Eq. 2 by using a previous result<sup>4</sup> that  $D_Z = (0.35 \pm 0.04)Ja^2$ .

The results are that  $D_E$  is temperature independent and proportional to  $J$  with a slope such that  $D_E = (0.75 \pm 0.15)Ja^2$ , where  $a$  is the nearest-neighbor distance.

Spin-diffusion constants have been predicted by several authors,<sup>9</sup> but no mention was made of the two energy transports. Redfield and Yu<sup>10</sup> recently calculated both  $D_Z$  and  $D_E$  in the high-temperature limit by the moment method. Their results are in essential agreement with previous calculations of  $D_Z$  and indicate that  $D_E$  is  $0.67Ja^2$ , about twice  $D_Z$ , for the bcc lattice. Thus there is good agreement

within the limits of the experiment and uncertainty in the theory.

We wish to express our thanks to Dr. A. G. Redfield for his suggestions and for permitting us to present some of his unpublished results. We are also deeply indebted to Dr. Horst Meyer for his constructive remarks on this manuscript.

†Research supported by a grant from the National Science Foundation and from the Army Research Office (Durham).

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<sup>1</sup>A. G. Redfield, private communication.

<sup>2</sup>For a discussion of the "three-bath model" in He<sup>3</sup>, see, for example, R. L. Garwin and A. Landesman, Phys. Rev. **133**, A1503 (1964).

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<sup>4</sup>J. R. Thompson, E. R. Hunt, and H. Meyer, Phys. Letters **25A**, 313 (1967). In this paper  $D_Z$  is denoted by  $D$ .

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<sup>9</sup>H. S. Bennet and P. C. Martin, Phys. Rev. **138**, A608 (1965); H. Mori and K. Kawasaki, Progr. Theoret. Phys. (Kyoto) **27**, 529 (1962); P. G. de Gennes, J. Phys. Chem. Solids **4**, 223 (1958); M. DeLeener and P. Resibois, Phys. Rev. **152**, 318 (1966).

<sup>10</sup>A. G. Redfield and W. N. Yu, to be published.  $D_E$  is calculated by substituting  $E_i, E_j$  for  $S_{iz}$ , where  $E_i$  is the sum of  $J\vec{S}_i \cdot \vec{S}_k$  over all nearest neighbors  $k$  of the  $i$ th spin, in all equations of A. Redfield, Phys. Rev. **116**, 315 (1959).

## SOUND PROPAGATION THROUGH A DILUTE SOLUTION OF He<sup>3</sup>-He<sup>4</sup> †

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(Received 15 December 1967)

In this communication, we present recent measurements of both the temperature and frequency dependence of the velocity of sound propagated through a 5.5 mole% solution of He<sup>3</sup> in He<sup>4</sup>. Attenuation measurements, which have been extended to lower temperatures than were obtained for those previously reported, are also presented.<sup>1</sup> In marked contrast to the behavior observed in He<sup>4</sup>,<sup>2</sup> the sonic velocity

in the solution is a strong function of the temperature even at the lowest temperature (35 mdeg); furthermore, the slope of the velocity-temperature curve up to ca. 300 mdeg is in the opposite sense to that observed in He<sup>4</sup>, i.e., the velocity of sound in the solution decreases with increasing temperature. Also, the attenuation at the low-temperature end goes as a high power of the temperature rather than