

$H \neq 0$ ) can be created through the use of second or higher Born approximations. Because of the extreme smallness of the scattering length as compared to the de Broglie wavelength of the neutron, it is reasonable to expect that the cross section for emitting any transverse wave should be much smaller than the corresponding cross section for a longitudinal wave. Thus, the existing measurements of total cross sections<sup>2,4</sup> already give an indication that these excitations are longitudinal waves. However, because of our limited knowledge concerning the unusual behavior of He II, it is desirable to have a direct experimental determination of the helicity of these excitations as discussed above. If experimentally one finds that both the long-wavelength ( $k < 1 \text{ \AA}^{-1}$ ) excitations and the short-wavelength ones ( $k \sim 2 \text{ \AA}^{-1}$ ) have zero as their longitudinal angular momentum (helicity) quantum number, then perhaps it would be clarifying to use the same name "phonon" for all of these excitations and regard the variation of  $E(k)$  as just a special form of dispersion relation.

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## FREE INDUCTION DECAYS OF ROTATING SOLIDS\*

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Motional narrowing and invariance of the second moment to motional narrowing have been observed for the  $F^{19}$  nuclear resonance line shapes in powdered  $CaF_2$  and Teflon due to rapid rotation of the sample.<sup>1</sup>

In the usual nuclear resonance experiment one

finds that internal motion within the sample is evidenced by a decrease in the sample's observed nuclear resonance line width and its second moment.<sup>2</sup> However, from the Van Vleck<sup>3</sup> expression for the second moment, one expects the second moment to be invariant to internal motion or rotation of the sample since the motional part of the Hamiltonian for the sample commutes with the total spin operator. It can be shown that internal motion increases the fourth moment,<sup>4</sup> which implies that the central portion of the resonance line narrows while the wings increase. The motional modulation of the resonance line produces side bands which add to the wings of the line. The motional contribution of the wings is too weak to be observed because the internal motion has a broad frequency spectrum and thus the side band contribution to the wings is spread over a broad frequency range. However, the nuclear magnetic resonance line shape for a solid sample rotating at a constant angular speed  $\omega_s$  should show sharp side bands at frequencies related to  $\omega_s$ .

Consider a solid containing a single magnetically active nuclear species placed in a uniform magnetic field  $H_0 \hat{z}$ . If the magnetic interaction among the nuclei is purely dipolar, the Hamiltonian<sup>3</sup> describing the solid is (ignoring internal motion and nonsecular terms)

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_0 + \mathcal{H}_1, \\ \mathcal{H}_0 &= -\gamma \hbar H_0 S_z, \\ \mathcal{H}_1 &= \frac{1}{2} \sum_{j \neq k} (A_{jk} \vec{s}_j \cdot \vec{s}_k + B_{jk} s_{jz} s_{kz}), \end{aligned} \quad (1)$$

$$B_{jk} = -3A_{jk} = -\frac{3}{2} (\gamma^2 \hbar^2 / r_{jk}^3) (3 \cos^2 \theta_{jk} - 1);$$

$\vec{r}_{jk}$  is the vector connecting the  $j$ th and  $k$ th nuclei,  $\theta_{jk}$  is the angle  $\vec{r}_{jk}$  makes with the  $z$  axis, and  $\gamma$  is the gyromagnetic ratio of the nuclear species.

If the sample is rotating about an axis oriented at an angle  $\theta_H$  with respect to  $H_0 \hat{z}$ , the addition theorem for Legendre polynomials yields

$$\begin{aligned} (3 \cos^2 \theta_{jk} - 1) &= \frac{1}{2} (3 \cos^2 \theta_H - 1) (3 \cos^2 \theta_{jk}' - 1) \\ &+ \frac{3}{2} \sin 2\theta_H \sin 2\theta_{jk}' \cos(\phi_{jk}' - \omega_s t) \\ &+ \frac{3}{2} \sin^2 \theta_H \sin^2 \theta_{jk}' \cos[2(\phi_{jk}' - \omega_s t)], \end{aligned} \quad (2)$$

where  $\theta_{jk}'$  is the angle between  $\vec{r}_{jk}$  and the axis of rotation. Calculations using the Hamiltonian derived from (1) and (2) are hampered by  $\mathcal{H}_1$  not commuting with itself at all times. Using the technique of Lowe and Norberg,<sup>5</sup> the free induction decay and thus the line shape can be calcu-

lated for the case of  $A_{jk} = 0$  for all  $j$  and  $k$ . The results are that

$$\langle \mu_x(t) \rangle = \frac{\mu_0 \cos \omega_0 t}{N_0} \sum_{j=1}^{N_0} \prod_k \cos \left( \int_0^t B_{jk}(t') dt' / 2\hbar \right), \quad (3)$$

the integration in Eq. (3) being trivial. For  $\theta_H = 0$ ,  $\langle \mu_x(t) \rangle$  is independent of the spinning speed of the sample. The inclusion of  $A_{jk}$  in the calculation will not change this result. For  $\theta_H = \arccos(1/\sqrt{3}) = 54.7^\circ$ ,  $\langle \mu_x(t) \rangle$  is a periodic function with a frequency  $f_S$ . One might facetiously call the extra signals "spinning echoes." The corresponding line shape has side bands at  $f_S$  and multiples of  $f_S$ . The inclusion of  $A_{jk}$  in the calculation for  $\theta_H = 54.7^\circ$  can be shown to make the amplitude of the spinning echoes decay, but it does not wash them out completely. For  $\theta_H = 90^\circ$ , the line shape displays side bands at  $2f_S$  and multiples of  $2f_S$ . Excluding the side band contribution to the line shape, the calculations show the line shape to be narrowed by a factor of two. The inclusion of  $A_{jk}$  in the calculation is again expected to change the amplitude of the side bands, but not their position.

Line shape measurements on spinning samples were made by observing the free induction decays of the spinning samples using a standard spin-echo apparatus.<sup>6</sup> The apparatus was operated at 30 Mc/sec, had a crossed coil rf head and a square-law detector and produced rf pulses lasting 4  $\mu$ sec that were able to nutate the  $F^{19}$  nuclei of the sample by  $90^\circ$ . The applied magnetic field  $H_0$  was determined to be homogeneous enough that it did not contribute noticeably to the nuclear magnetic resonance line shape of the solid. The samples were spun at speeds up to 7 kc by using them as rotors of an air-driven turbine. The samples were cylinders 7 mm wide, 7 mm in diameter, and had nylon bearings, and spun on phosphor bronze axles. The spinning frequency was limited only by the strength of the sample. The orientation of the spinning axis could be varied with respect to the applied magnetic field  $H_0$ . The curves displayed in Figs. 1 and 2 are the free induction decay shapes for spinning and nonspinning samples, and their corresponding Fourier transforms (which are the cw, nuclear magnetic resonance line shapes<sup>5</sup>). It may be noted first that to within the resolution of the drawings (and the original data), the free induction decays have the same initial second derivative. Thus the corresponding cw line shapes have the same second moment, and the

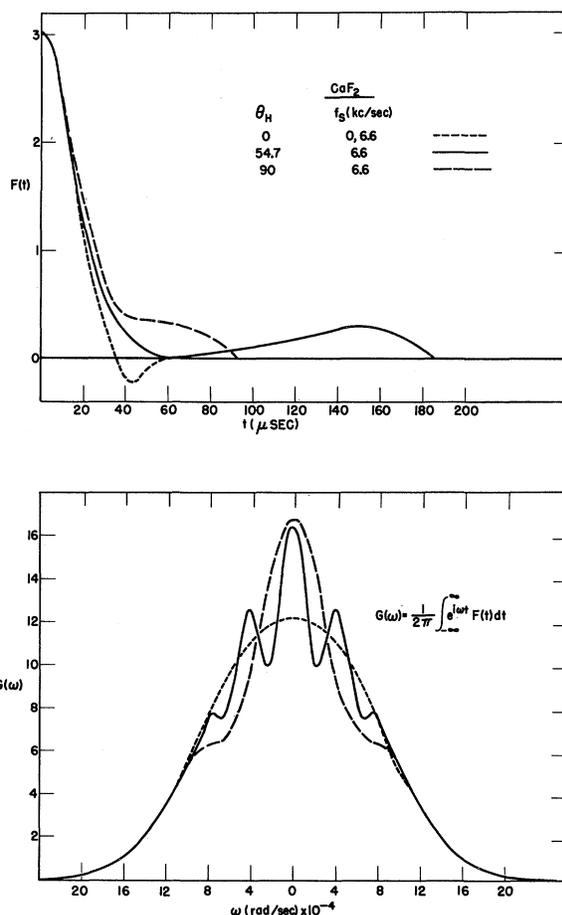


FIG. 1. Free induction decays for spinning and nonspinning samples of powdered  $CaF_2$  and their Fourier transforms. The curves are corrected for instrumental nonlinearities.

second moment is invariant to motional narrowing in this case. It is further observed that when  $\theta_H = 0$ , the free induction decay and line shape are the same for the spinning and nonspinning case. When  $\theta_H = 54.7^\circ$ , the free induction decays show a series of peaks or "spinning echoes" at  $T_S$ ,  $2T_S$ , etc., and the cw line shape's central portion narrows while the intensity in the wings increases at approximately  $\omega_S$ ,  $2\omega_S$ , etc. For  $\theta_H = 90^\circ$ , the free induction decay is seen to lengthen and the central portion of the cw line shape to narrow correspondingly. One may reasonably interpret the shoulders on the cw line shape to be due to the side band contribution at  $2\omega_S$ .

With further refinement of this spinning technique and its theory, one might use it to study nondipolar contributions to line shapes in solids.

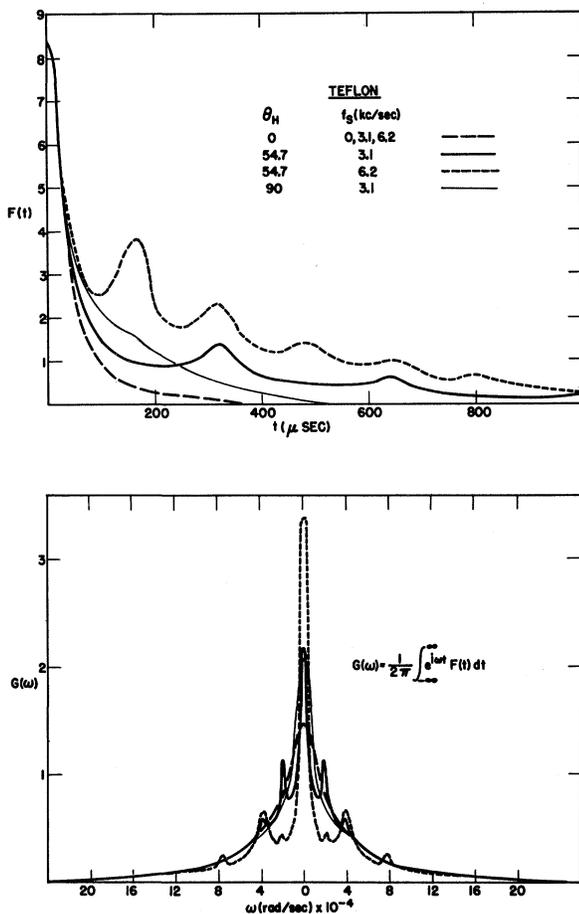


FIG. 2. Free induction decays for spinning and nonspinning samples of Teflon and their Fourier transform. The curves are corrected for instrumental nonlinearities.

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<sup>1</sup> Such effects have been independently observed for the Na<sup>23</sup> nuclear resonance in NaCl by Andrews, Bradbury, and Eades, Arch. sci. Geneva **11**, 223 (1958).

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## DIFFUSIVE MOTIONS IN LIQUIDS AND NEUTRON SCATTERING

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Measurements of neutron inelastic scattering by a classical liquid provide access to the details of the atomic motions in the liquid, by way of time-dependent correlation functions introduced by Van Hove.<sup>1</sup> The author has discussed some features of these correlation functions in connection with a series of experiments<sup>2</sup> on the isotopic pair—light water which scatters incoherently, and heavy water which scatters coherently. These functions have also been discussed by Vineyard.<sup>3</sup> The partial differential neutron scattering cross sections are Fourier transforms over space and time of the time-dependent correlations,<sup>1</sup> and essentially are functions only of neutron energy transfer and momentum transfer, i.e., of  $\omega = \hbar^{-1}(E_0 - E')$  and  $\vec{Q} = \vec{k}_0 - \vec{k}'$ , where  $E_0$ ,  $E'$ ,  $\vec{k}_0$ ,  $\vec{k}'$  are, respectively, the ingoing and outgoing neutron energies and wave vectors. The coherent cross sections are determined by the pair correlation function, which classically is “the probability that, given an atom at position zero at time zero, any atom is at position  $\vec{r}$  at time  $t$ .” The incoherent cross sections are determined by the self-correlation function, which classically is “the probability that, given an atom at position zero at time zero, the same atom is at position  $\vec{r}$  at time  $t$ .”

The experiments<sup>2</sup> led to the conclusion that the qualitative behavior of the cross sections is largely determined by the asymptotic behavior of the correlation functions at very small times and at large times. Because the correlation functions for any system (solid, liquid, or gas) change very rapidly at small times, the scattering contains an inelastic component which, for a classical system at a given large  $Q$ , is almost independent of the state of the system, being determined by the temperature and by the mass of the