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OBSERVATION OF THE λ ANOMALY IN SOLID D_2 BY NUCLEAR MAGNETIC RESONANCE*

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Several unsuccessful attempts have been made by nuclear magnetic resonance techniques[1-3] to observe a cooperative transition in solid n-D_2 (33% para) analogous to the λ-type anomaly in solid n-H_2 (75% ortho). In addition, Smith and Housley[4] found no transition even in enriched samples of D_2 with para D_2 concentrations up to 55%. The transition in solid H_2 occurs due to the quenching of the rotational motion of the ortho H_2 ($I=1$, $J=1$) molecules implying that the transition temperature T_λ is a sensitive function of the ortho H_2 concentration[5,6] and the density.[3-7] Recent heat capacity measurements by Grenier and White[8] demonstrated the existence of such a cooperative transition in solid D_2 above 1.5°K providing the para D_2 ($I=1$, $J=1$) concentration was in excess of 60%. We have made nuclear-resonance studies of enriched para D_2 samples (with concentration of about 80%) and have observed the splitting of the resonance line in D_2 below 2°K.

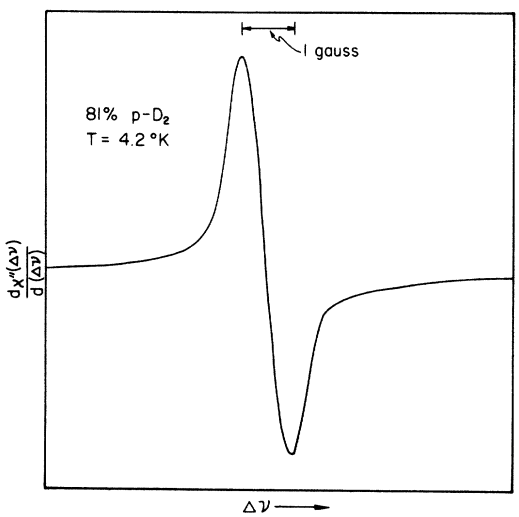

FIG. 1. The derivative with respect to frequency of the absorption signal from an 81% pD_2 sample at 4.2°K.

The enriched para D_2 samples were prepared in a manner similar to that suggested by Cunningham, Chapin, and Johnston.[9] The specimen was condensed from the gas phase into a Teflon sample cell surrounded by a small amount of liquid H_2 and then frozen quickly by transferring liquid He on top of the H_2. Figure 1 shows a derivative of the resonance line at 4.2°K obtained with a sample containing 81% p-D_2. This line is actually a superposition of lines arising from molecules with $I=2$ (ortho D_2) and $I=1$ (para D_2). Since the molecules with $I=2$ have $J=0$ at these low temperatures, one does not expect that quenching of rotation will have any effect on their contribution to the resonance line, in a first approximation. This trace was made with a frequency sweep rate of 45 cps/sec at a Larmor frequency of 2.63 Mc/sec. The width of the line shown is approximately 1.0 gauss as compared to the value of about 1.5 gauss obtained in n-D_2.[3-10] The asymmetry of the line becomes more pronounced as the frequency sweep rate is increased.

Reduction of the temperature to about 2.2°K revealed the existence of side peaks split apart by 76.8 ± 0.5 kc/sec. Figure 2 shows a trace of the resonance line at 1.2°K with 100 kc/sec reference marks. The magnitude of the splitting calculated by Reif and Purcell[2] is

$$\Delta\nu = 3\tau d,$$

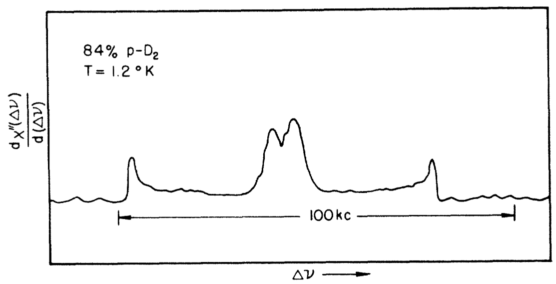

FIG. 2. The derivative of the resonance line at 1.2°K after the splitting has occurred.

where d for the free D_2 molecule has the value[11] 25.24 kc/sec. For D_2, d is a function of the strength of the dipolar interaction and the magnitude of the deuteron nuclear quadrupole moment. The calculated value of τ is 1.01,[2] the 1% correction term arising from the contributions of higher order spherical harmonics in the perturbation calculation. Thus the calculated value of the splitting ($\Delta\nu_{\text{calc.}} = 76.5$ kc/sec) is in excellent agreement with the experimental value.

After the splitting of the line occurred, several other effects were observed: (i) The signal-to-noise ratio deteriorated rather badly; (ii) the line became so asymmetric that the recorder trace bore little resemblance to the conventional derivative curve; (iii) the remnant central line became noticeably broader, with a width of 5-8 gauss. All attempts to recover the symmetry of the line (such as reduction of the sweep rate and lowering the radiofrequency power) failed. Investigations into the origin of the asymmetry and the apparent loss of signal after the λ transition are in progress.

Due to the relatively low signal-to-noise ratio, no attempt was made at this time to determine the transition temperature from the nmr data. The value of the transition temperature for 81% p-D_2 derived from the heat capacity data is about 2.8°K.

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NONLINEAR ANISOTROPIC TERMS IN THE FREE-CARRIER FARADAY ROTATION IN CUBIC SEMICONDUCTORS

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The nonlinearity of the magnetic field dependence of free-carrier Faraday rotation in cubic semiconductors has been investigated theoretically and experimentally in the low-field region ($\mu B \ll 1$, where μ and B are the mobility and the magnetic field, respectively). The effect arises due to B^3 terms in the expansion of the high-frequency electric current in the presence of the magnetic field. Unlike the first-order term of the Faraday rotation, the nonlinearity is anisotropic for nonspherical energy bands. The effect is particularly interesting in the low-frequency range ($\omega\tau \ll 1$, ω and τ being the frequency and the collision time, respectively), where it provides a direct measure of the scattering average $S \equiv \langle\tau^4\rangle/(\langle\tau^2\rangle\langle\tau\rangle^2)$. Measurements of the nonlinear terms were carried out at room temperature at 35 Gc/sec on a series of n-type silicon crystals. The observed anisotropy of the effect agreed well with the theoretical analy-