helium-cooled surfaces. A liquid-nitrogen-cooled radiation shield surrounded the sample rod and helium reservoir except for the slots used for introducing the molecular beam and for pumping.

For resonance measurements, the sample-coated sapphire rod was inserted into a rectangular microwave cavity through an aperture 4 mm in diameter. The cavity was operated in the *TE* 012 mode and kept at liquid nitrogen temperature by thermal contact with the radiation shield. The penetration of the sample rod into the cavity was adjustable up to a maximum of 16 mm. The broad face of the cavity was perpendicular to the magnetic field supplied by a 12-in. Varian magnet. The microwave frequency of the klystron was stabilized at the sample cavity resonance frequency of about 9066 Mc/sec. The spin resonance was observed by slowly sweeping the magnetic field through resonance and recording the absorption signal derived from the field modulation at 400 cps. The magnetic field was measured by a proton resonance magnetometer. The microwave frequency was measured by using a standard compared with radio station WWV.

Observations of the electron spin resonances in hydrogen and deuterium samples have shown that the trapped atoms are quite stable at liquid helium temperature. The resonance lines appeared immediately after deposition and remained detectable as long as liquid helium temperature was maintained.

The spectrum for hydrogen consisted of two lines of almost equal intensity, one 234.1 oersteds above and the other 274.6 oersteds below the field for free-electron resonance. The half-width for each component was about 0.7 oersted. The observed spectrum for deuterium was a triplet with the center line three to four times stronger than either of the other components. The center line was 2.1 oersteds lower than the free-electron resonance field and was separated from the low- and high-field lines by 76.7 and 78.7 oersteds, respectively. The half-width of the deuterium lines was roughly 1.3 oersteds.

The spectral patterns and line intervals for the observed hydrogen doublet and deuterium triplet are consistent with the known hyperfine structures of free hydrogen and deuterium atoms. By applying the Breit-Rabi formulas,[1,2] we have obtained the hyperfine coupling constants and $g_J$ values for trapped atoms shown in Table I. Comparing these values with the values for free atoms, also shown in Table I, it is seen that the atoms trapped by low-temperature deposition are very nearly, but not exactly, free. We are not ready at this time to explain quantitatively the small deviations from the free states, the difference between the hydrogen and deuterium line widths and, especially, the anomalous intensity of the deuterium center line.

TABLE I. Hyperfine splitting constants ($\nu_c$) and $g_J$ values for H and D atoms.

| | H | D |
|---|---|---|
| $\nu_c$ (trapped) Mc/sec | 1417.13±0.45 | 326.60±0.23 |
| $\nu_c$ (free) Mc/sec[a] | 1420.4057 | 327.3843 |
| Deviation of $\nu_c$ from free value | 0.23% | 0.24% |
| $g_J$ (trapped) | 2.00243±0.00008 | 2.00244±0.00008 |
| $g_J$ (free)[b] | 2.00230 | |

[a] P. Kusch, Phys. Rev. **100**, 1188 (1955).
[b] R. Beringer and M. A. Heald, Phys. Rev. **95**, 1474 (1954).

It was also observed that each hyperfine line of the hydrogen atom was accompanied by two satellite lines symmetrically spaced about it. The separation of these satellites from their main line corresponds to the energy required to flip a neighboring proton.[3] Similar satellites have also been observed for each of the deuterium lines except that they were not well resolved from the relatively broad deuterium line.

In the course of our experiments with hydrogen, a number of strong, sharp lines of unknown origin have been observed in the vicinity of the free electron resonance field. These appeared as triplet and quartet spectral groups, each having uniformly spaced lines. The line spacing varied from 4 to 23 oersteds among the groups. It is possible that these lines are connected with excited molecules stabilized in the low-temperature matrix.

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[2] J. E. Nafe and E. B. Nelson, Phys. Rev. **73**, 718 (1948).
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## Antiferromagnetic Resonance in $MnF_2$†

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ANTIFERROMAGNETIC resonance has been observed in a single crystal of $MnF_2$.[1] Observations on this absorption line have been made in the temperature range from 36° to 65°K and at frequencies from 213 300 Mc/sec to 96 000 Mc/sec.[2] $MnF_2$ has tetragonal symmetry and a Néel temperature of approximately 68°K. The interpretations of antiferromagnetic resonance experiments on other crystals[3-6] have been complicated by a lower order of crystal symmetry, low Néel temperatures, or the necessity of working very close to the Néel temperature. In contrast, $MnF_2$ furnishes the most direct and simple test of the basic theory.

It has been shown by Keffer and Kittel[7] and others that the resonance frequency with the external magnetic field applied parallel to the symmetry axis is given by:

$$\omega/\gamma=(2H_EH_A)^{\frac{1}{2}}\pm H_0(1-\alpha/2), \tag{1}$$

where $H_E$ is the exchange field, $H_A$ is the anisotropy field, $H_0$ is the applied field, $\gamma$ is $ge/2mc$, and $\alpha$ is the ratio of the parallel and perpendicular susceptibilities.

In addition, Keffer[8] has estimated $H_E$ and $H_A$ for $MnF_2$ at 0°K. He calculated the anisotropy field as 8800 gauss, of which 8300 gauss arise from magnetic dipole interactions and 500 gauss from interactions with the crystalline electric field. From molecular field theory and susceptibility measurements,[9] $H_E$ is 540 000 gauss. This yields a value for $(2H_EH_A)^{\frac{1}{2}}$ of 97 500 gauss. At higher temperatures, Keffer assumed that $(2H_EH_A)^{\frac{1}{2}}$ varies as $M/M_0$, which in turn is assumed to vary as the modified Brillouin function for $S=5/2$.

In the present experiment, millimeter wave power from a silicon crystal harmonic generator is sent through either a 0.016-in. or a 0.033-in. thick $MnF_2$ slab mounted in a 0.122×0.061-in. wave guide with the symmetry axis of the crystal parallel to the dc magnetic field. The transmitted power is monitored by a silicon crystal detector as the magnetic field is changed from zero to about 9000 gauss. One measurement was made with the third harmonic of a 4.219-mm pulsed magnetron.[10] The experimental observations are given in Table I, where $\Delta H$ is the full half-width uncorrected for $(1-\alpha/2)$.

TABLE I. The temperature, line width, $g$ value, and percent absorption are given for the various frequencies at which the antiferromagnetic resonance was observed, at zero magnetic field.

| $\nu$ (kMc/sec) | $T$ (°K) at $H=0$ | $\Delta H$ (gauss) | $g$ | % absorption for 0.016-in. slab |
|---|---|---|---|---|
| 96 | 63.6±0.5 | ~8000 | | ~15 |
| 117 | 62.3±0.8 | ~7000 | | ~50 |
| 146 | 57.5±0.6 | ~5500 | 1.9±0.6 | |
| 213.3 | 42.0±0.4 | 1600 | 2.1±0.15 | ~95 |

If it is assumed with Keffer that $\nu$ varies as the modified Brillouin function, then

$$\nu_{H=0}=KB_{\frac{5}{2}}(T/T_N). \qquad (2)$$

It was decided to use 67.7°K as $T_N$. This value was obtained from the susceptibility measurements[9] and gives a good fit with the experimental data. In addition, it is consistent with the fact that traces of paramagnetic resonance absorption[11] have been detected at temperatures as low as 67.6°K. The point at 213.3 kMc/sec is then sufficient to determine $K$ as 264±5 kMc/sec. Since $g$ was determined to be 2.1±0.15, $(2H_EH_A)^{\frac{1}{2}}$ is equal to 90 000±8000 gauss, in very good agreement with Keffer's estimate of 97 500 gauss.

The experimental points are shown in Fig. 1. Also shown is the curve given by Eq. (2) with $K=264$ kMc/sec and $T_N=67.7$°K.

Qualitative observations were made as various angles were changed. When the crystal was rotated at constant temperature and frequency with fixed mutually perpendicular dc and rf magnetic fields, the intensity of the absorption rapidly decreased as the symmetry axis was rotated away from the direction of the dc field. In addition, the magnetic field necessary for resonance increased. This increase in magnetic field was also observed as the dc magnetic field was rotated with the rf field and crystal fixed. These rotation experiments verified that the symmetry axis of the crystal coincided with the magnetic anisotropy axis, in agreement with the susceptibility measurements.

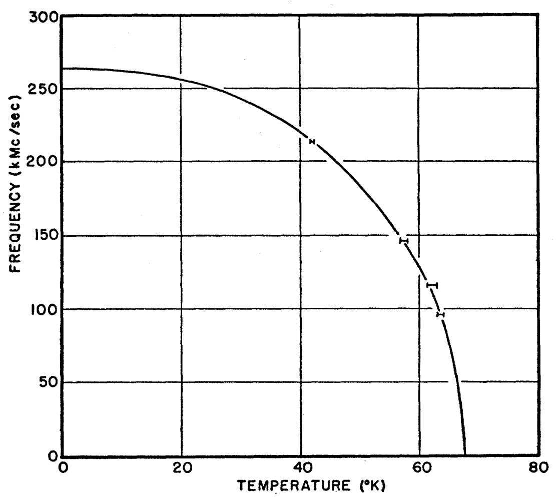

FIG. 1. Temperatures at which antiferromagnetic resonance occurs in $MnF_2$ at $H=0$ for various frequencies. Equation (2) with $K=264$ kMc/sec and $T_N=67.7$°K is also shown.

The $g$ value of $MnF_2$ in the paramagnetic state[12] at 300°K is 2.05±0.03. This agrees with the value for the antiferromagnetic state within its error. This is evidence that the factor $(1-\alpha/2)$, which at 42°K amounts to 0.75, is indeed correctly present in the theory.

The results of this experiment are in complete agreement with the general theory of antiferromagnetic resonance developed by Keffer and Kittel as well as with the detailed predictions of Keffer for $MnF_2$. However, if the observed frequencies are interpreted as being proportional to the magnetization of the lattice, they are in disagreement with the direct measurement of the magnetization by the method of neutron scattering.[13] The reasons for this discrepancy are not evident at present.

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[1] This single crystal was kindly provided by Professor J. W. Stout. We are also indebted to Professor Stout for a discussion of our results.

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[7] F. Keffer and C. Kittel, Phys. Rev. **85**, 329 (1952).

[8] F. Keffer, Phys. Rev. **87**, 608 (1952).

[9] M. Griffel and J. W. Stout, J. Chem. Phys. **18**, 1455 (1950).

[10] This magnetron was kindly made available to us by Mr. M. J. Bernstein of this laboratory.

[11] C. A. Hutchinson (private communication).

[12] We wish to thank Professor G. K. Fraenkel and Dr. B. Segal for kindly measuring this quantity.

[13] R. A. Erickson, Phys. Rev. **90**, 779 (1953).