

finite as the temperature  $T$  approaches the critical temperature  $T_c$  from below. Therefore when  $T$  is sufficiently near  $T_c$  and  $d$  is finite,  $\lambda$  is much greater than  $d$  and so Eq. (8) applies. This shows that the unit of flux quantization tends to zero as  $T$  tends to  $T_c$ . Since  $|\vec{J}|$  is proportional to  $\Phi$  it also tends to zero and so does the density  $|\Psi|^2$ . However, the ratio  $|\vec{J}|/|\Psi|^2$ , which occurs in the fluxoid, tends to  $ne\hbar/mv$ .

When the tube is not thin compared to  $\lambda$ , we have obtained the quantum of flux through the hole as well as the magnetic field as a function of radial distance from the axis by solving Maxwell's equations for  $\vec{A}$  with  $\vec{J}$  given by Eq. (1). By using these results we have computed the Ginzburg-Landau expression for the free energy of the equilibrium states. It is the sum of a surface energy proportional to  $n^2$  and the free energy in the absence of a magnetic field, which is independent of the quantum number  $n$ . When  $n$  is small and the tube is thick compared to  $\lambda$ , the surface energy is negligible. In any case these equilibrium values must be interpreted as local minima with respect to small deviations from equilibrium.

We wish to thank Professor C. N. Yang for a stimulating discussion of his microscopic theory.

- <sup>1</sup>F. London, *Superfluids* (John Wiley & Sons, Inc., New York, 1950), Vol. 1, p. 152.
- <sup>2</sup>M. R. Schafroth, Phys. Rev. **96**, 1149, 1442 (1954).
- <sup>3</sup>L. N. Cooper, Phys. Rev. **104**, 1189 (1956).
- <sup>4</sup>L. Onsager (private communication); see also Phys. Rev. Letters **7**, 50 (1961).
- <sup>5</sup>B. S. Deaver and W. M. Fairbank, Phys. Rev. Letters **7**, 43 (1961).
- <sup>6</sup>R. Doll and M. N abauer, Phys. Rev. Letters **7**, 51 (1961).
- <sup>7</sup>N. Byers and C. N. Yang, Phys. Rev. Letters **7**, 46 (1961).
- <sup>8</sup>J. M. Blatt, Phys. Rev. Letters **7**, 82 (1961).
- <sup>9</sup>J. Bardeen, this issue [Phys. Rev. Letters **7**, 162 (1961)].
- <sup>10</sup>L. P. Gor'kov, J. Exptl. Theoret. Phys. (U.S.S.R.) **36**, 1918 (1959) [translation: Soviet Phys. - JETP **9**, 1364 (1959)].
- <sup>11</sup>V. L. Ginzburg and L. D. Landau, J. Exptl. Theoret. Phys. (U.S.S.R.) **20**, 1064 (1950). See also V. L. Ginzburg, Nuovo cimento **2**, 1234 (1955).
- <sup>12</sup>J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

## ELECTRICALLY INDUCED SHIFT OF THE $F^{19}$ RESONANCE FREQUENCY IN $MnF_2$ <sup>†</sup>

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The predicted linear effect of an applied electric field on the magnetic hyperfine interaction<sup>1</sup> has been found experimentally in antiferromagnetic  $MnF_2$ . We believe this is the first such observation, although the quadratic variation with  $E$  of the hyperfine splitting in the Cs atom has been reported earlier.<sup>2</sup>

The  $F^{19}$  resonance,<sup>3</sup> which occurs at 159.970 Mc/sec without external magnetic field at 4.2°K, was detected with the spectrometer built by Kushida for the investigation of the pressure dependence of the same resonance.<sup>4</sup>

A plane parallel slab, 1 mm × 5 mm × 20 mm, was cut from a single crystal of  $MnF_2$  with the [100] direction perpendicular to the broad face. Metal electrodes were applied to the broad face by silver paint that was fired at 1000°F. Contacts were then attached with indium solder. A dc voltage up to 9700 volts could be applied from a battery of dry cells. It was necessary to use rf chokes to

isolate the high-voltage line from the tank circuit. At the highest field,  $E = 0.97 \times 10^5$  volts/cm, the  $F^{19}$  resonance is completely resolved into two components. In Fig. 1 the recordings of the absorption derivative are reproduced for zero field and three values of the applied electric field. They are compatible with a linear absolute shift  $\Delta\nu_E = 5$  kc/sec for each of the four  $F^{19}$  nuclei in the unit cell, if  $E = 10^4$  volts/cm is applied in the [100] direction, as shown in Fig. 2. The separation of the two components is of course twice the shift.

For an arbitrary direction of  $E$  the line should split into four components. Only the component of  $\vec{E}$  pointing from each  $F^{19}$  nucleus to the nearest  $Mn^{++}$  ion is effective in shifting the resonance of that nucleus. If  $E = 10^4$  volts/cm is applied along a [110] direction, half the  $F^{19}$  nuclei will keep an undisplaced resonance, while one quarter will be displaced by 7.1 kc/sec and the remaining quarter by -7.1 kc/sec.

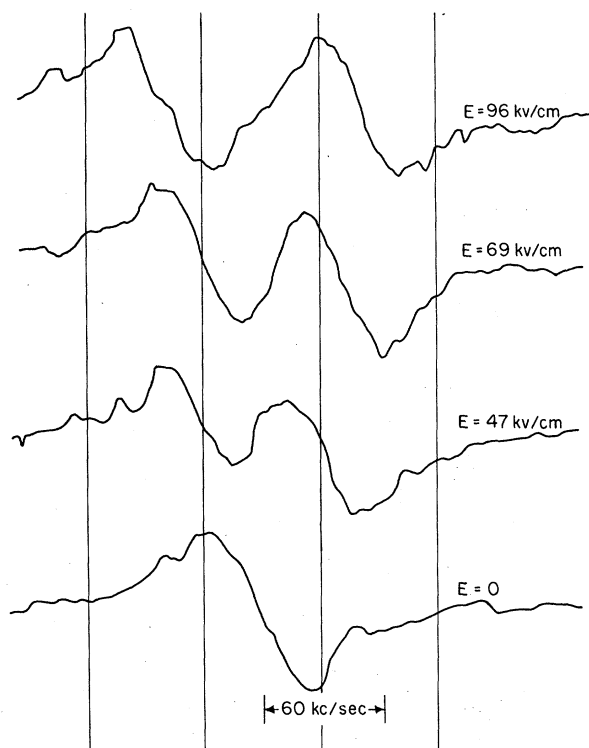


FIG. 1. The absorption derivation of the  $F^{19}$  resonance in  $MnF_2$  at  $4.2^\circ K$  in zero magnetic field. An electric field of variable strength is applied along the [100] crystallographic axis.

This experimental result is an order of magnitude smaller than the admittedly crude theoretical estimate,<sup>1</sup> which was based on the assumption of an isotropic binding force of the  $F^-$  ion in the (001) plane. Inspection of the nuclear distances in  $MnF_2$  and the ionic radii of  $F^-$  and  $Mn^{++}$  reveals that the  $F^-$  ion at the position  $(u, u, 0)$  is most tightly bound with the  $Mn^{++}$  ion at the origin, and somewhat weaker with the  $Mn^{++}$  ions at  $(\frac{1}{2}, \frac{1}{2}, \pm \frac{1}{2})$ . Its interaction with  $F^-$  ions and farther  $Mn^{++}$  ions should be much weaker. The binding is therefore highly anisotropic. When one uses the known force constants of the  $MnF$  and  $MnF_2$  molecules,<sup>5</sup> the change in the nearest Mn-F distance  $\Delta r_{II}$  could be nearly an order of magnitude less than estimated earlier. The assumption that  $u$  does not change with hydrostatic pressure may also have to be revised. If the internuclear distance  $r_{II}$  is most difficult to change,  $u$  would increase somewhat under compression. This would make the values for  $(\partial A / \partial r)_{I, II}$  quoted by Benedek and Kushida<sup>4</sup> too large. A revision of the covalent effects, utilizing the refinements pointed out by

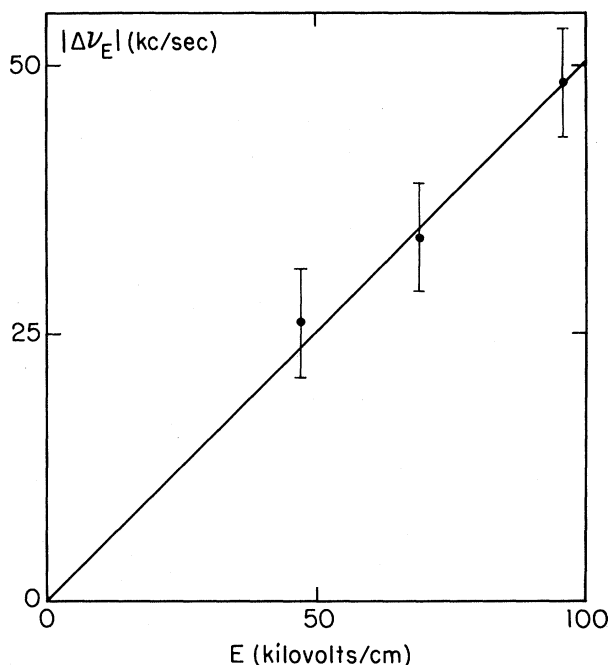


FIG. 2. The absolute shift of the  $F^{19}$  resonance versus applied electric field strength along [100]. The "electric doublet splitting" is twice the shift and amounts to 1 cps/(volt/cm).

Freeman and Watson, and a determination of the internal electric field at the bond position, is also necessary to obtain agreement with experimental observation.

Finally it is pointed out that the observed effect may be used to study the presence or absence of antiferromagnetic domain walls. Néel introduced this concept, and several interesting theoretical discussions have appeared,<sup>8,9</sup> but experimental evidence is difficult to obtain. Consider a crystal of  $MnF_2$  with an external magnetic field  $H_0$  ( $> 25$  gauss) along the  $c$  axis, [001]. Two  $F^{19}$  magnetic resonance lines will be observed. Apply an electric field along the [110] direction. If the crystal is a single domain, only one component of the "magnetic doublet" will be split further. Work on this problem is in progress.

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<sup>1</sup>N. Bloembergen, Phys. Rev. Letters **7**, 90 (1961).

<sup>2</sup>R. D. Haun and J. R. Zacharias, Phys. Rev. **107**, 107 (1957).

<sup>3</sup>V. Jaccarino and R. G. Shulman, Phys. Rev. **107**, 1196 (1957).

<sup>4</sup>G. B. Benedek and T. Kushida, Phys. Rev. **118**, 46 (1960).

<sup>5</sup>G. D. Rochester and E. Olsson, *Z. Physik* **114**, 495 (1939).

<sup>6</sup>A. J. Freeman and R. E. Watson, *Phys. Rev. Letters* **6**, 343 (1961); also *Phys. Rev.* (to be published).

<sup>7</sup>L. Néel, *Proceedings of the International Conference*

*on Theoretical Physics, Kyoto, 1953* (Science Council of Japan, Tokyo, 1954), pp. 711-714.

<sup>8</sup>Y. Y. Li, *Phys. Rev.* **101**, 1450 (1956).

<sup>9</sup>J. W. Cahn and R. Kikuchi, *J. Phys. Chem. Solids* **20**, 94 (1961).

## COMPARISON OF THE BETA SPECTRA OF B<sup>12</sup> AND N<sup>12</sup>†

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The observed near equality of the vector beta coupling constant given by the decay of O<sup>14</sup> and the muon decay coupling constant led Feynman and Gell-Mann<sup>1</sup> to postulate the conservation of the vector part of the beta-decay current. Gell-Mann<sup>2</sup> pointed out that a consequence of this conserved vector current (CVC) theory is an additional coupling in the beta interaction analogous to the anomalous magnetic moment of the nucleon ("weak magnetism"). The interference between this term and the Gamow-Teller transition amplitude for a 1<sup>+</sup> → 0<sup>+</sup>, ΔT = 1 beta transition then results in a deviation from the allowed shape of the B<sup>12</sup> and N<sup>12</sup> spectra. The magnitude of the effect can be calculated<sup>2,3</sup> from the rate of the analogous M1 gamma transition in C<sup>12</sup>. If S(E) is the spectrum of each transition, divided by the corresponding allowed Fermi spectrum, then the CVC theory predicts

$$S(E, B^{12})/S(E, N^{12}) = \text{const}[1 + (A + \delta A)E]f(E),$$

where the constant A arises from this interference term. The quantity f(E) is the inner bremsstrahlung correction. The expected value for A is (1.33 ± 0.15)% per Mev with an electromagnetic correction<sup>4</sup> δA of (-0.25 ± 0.15)% per Mev. The uncertainty in A comes from the uncertainty in the C<sup>12</sup> gamma-decay rate and the uncertainty in δA is an estimate of the error from using the shell model. Without the CVC theory hypothesis, A is estimated to be roughly 5 times smaller.<sup>4,5</sup>

The beta spectra of B<sup>12</sup> (E<sub>max</sub> = 13.369 Mev) and N<sup>12</sup> (E<sub>max</sub> = 16.43 Mev) were analyzed with the iron-free single-lens magnetic spectrometer described by Hornyak et al.<sup>6</sup> The baffle system was modified for the high electron energies involved. B<sup>12</sup> (20.3 msec) was produced by bombarding targets of natural boron with 1.65-Mev deuterons, and N<sup>12</sup> (12.5 msec) was produced by bombarding targets of enriched (96%) B<sup>10</sup> with 2.75-Mev He<sup>3</sup> ions. Both beams were pulsed at 60 cps at the ion source of the electrostatic ac-

celerator. The targets consisted of approximately 0.3 mg/cm<sup>2</sup> of boron deposited by cracking diborane on foils of thickness 0.5 mg/cm<sup>2</sup> and 3 mg/cm<sup>2</sup> for B<sup>12</sup> and N<sup>12</sup>, respectively. The beta rays were detected in a 10-mm thick anthracene scintillator. This is sufficiently thin (approximately 2.3-Mev range for electrons) to give nearly identical pulse-height spectra for all energies in the range studied (5-13 Mev). The use of a thin scintillator allows amplifier gain and discriminator settings to be left constant, reduces background as a result of its small volume, and produces a negligible small pulse tail extending into the noise background.<sup>7</sup> The activity produced on the target was monitored by the reaction protons from B<sup>10</sup>(d, p)B<sup>11</sup> for B<sup>12</sup> and B<sup>10</sup>(He<sup>3</sup>, p)C<sup>12</sup> for N<sup>12</sup>. A silicon p-n junction counter mounted in the spectrometer near the target served as a proton detector.

The spectra were sampled at 0.5-Mev intervals in the range 5 to 10.5 Mev for B<sup>12</sup> and 5 to 13 Mev for N<sup>12</sup>, the data being collected at alternating high- and low-energy points to minimize effects of target deterioration and instrumental drifts. The lower limit of 5 Mev was dictated by uncertainties in the branching ratios to excited states of C<sup>12</sup>; the upper limit was maintained well below the spectrum end points to minimize uncertainties arising from calibration errors and background subtraction. The background was studied at energies above the end points of the beta spectra, at zero field, and by using target backings without the boron. The neutron background was separately investigated with a beryllium target. Typical background corrections at 8 Mev amounted to about 1.7% for B<sup>12</sup> and 4.2% for N<sup>12</sup>. A B<sup>12</sup> spectrum taken with the relatively thicker backing used for the N<sup>12</sup> showed no deviation from the other B<sup>12</sup> spectra.

The observed spectra are corrected for branching<sup>8,9</sup> to the 4.4- and 7.6-Mev excited states of C<sup>12</sup> and for inner bremsstrahlung.<sup>4</sup> The N<sup>12</sup> branch to the 7.6-Mev state is assumed to be 3.5%. The