given by Van Vleck<sup>33</sup>; in fact, there exists a similar, although not nearly so simple relationship between the two as that between the intensity of the sidebands and Van Vleck's form of the second moment.

It would be of interest to verify experimentally not only the narrowing of the center line but also the existence of the sidebands. The fact that this has not been possible so far is due to the relatively small intensity of the sidebands. In order to permit their individual

<sup>33</sup> See reference 3, Eq. (29).

observation, it is necessary that the sidebands are well separated from the center line and the condition of validity  $|f| \gg |a|$  of the theory, presented here, expresses this very circumstance. With  $\langle \Delta_1^2 \rangle(c)$  of the order of magnitude  $|a|^2$ , it follows from Eq. (105) that the intensity of well separated sidebands is necessarily small, compared to that of the center line, even in the most favorable case where  $\sin^2\theta = 1$ . Nevertheless, the observation may become possible if larger signal-tonoise ratios can be achieved.

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# Double Nuclear Magnetic Resonance and the Dipole Interaction in Solids\*

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The nuclear magnetic dipole interactions in a polycrystalline sample of sodium fluoride have been investigated using an extension of the magnetic resonance technique. The resonance absorption of Na<sup>23</sup> was observed with a variable-frequency spectrometer in a fixed external magnetic field, and the mean square width of the line shape computed.

This width has two main sources: (a) the interaction of Na<sup>23</sup> nuclei with other Na<sup>23</sup> nuclei through the dipole fields of the magnetic moments, and (b) the interaction of  $Na^{23}$  nuclei with neighboring  $F^{19}$  nuclei by the same mechanism. The application of a strong rf field satisfying the resonance condition for F<sup>19</sup> nuclei in the same external field caused rapid transitions between the fluorine Zeeman energy levels, and thus altered the average field produced at the position of sodium nuclei by the nuclear magnetic moments of fluorine. By varying the strength of this rf field and also by varying the deviation from precise resonance of the F<sup>19</sup> spins, one could selectively alter the contributions of this source of line broadening.

The available rf field was not sufficiently intense to eliminate entirely the broadening caused by source (b). However, the detailed behavior of this contribution to the width as a function of the deviation from resonance agrees well with a theory of Bloch for the case of the rf field strong compared to the line widths involved.

## I. INTRODUCTION

HE line widths characteristic of nuclear magnetic resonance absorption in solids are commonly of the order of kilocycles/second, owing to the magnetic dipole interaction between nuclei localized in the crystal lattice. In general, the rigorous calculation of the line shape itself is prohibitively difficult because of the extremely large number of spins to be considered in a crystal of practical size. Recently Lowe and Norberg have made some progress in developing such a theory.<sup>1</sup> Van Vleck, however, has given a procedure<sup>2</sup> for calculating the moments of the line shape, and the second moment, in particular, is a measure of the mean

square line width. In a crystal containing two magnetic ingredients, A and B, the second moment of the resonance line of A will have contributions from both the A-A and the A-B magnetic dipole interactions.

The present work concerns itself with a method of altering this natural line shape by applying a perturbation which affects the A-B dipolar coupling. The accompanying paper by Bloch<sup>3</sup> presents a theoretical treatment of the experiment.

The concept of "averaging out" local fields in nuclear resonance experiments is not new. An early example is the explanation by Bloembergen, Purcell, and Pound<sup>4</sup> of the narrow lines observed in liquid samples. By virtue of the rapid molecular tumbling motions, nuclei are caused to sample many different local fields in a time short compared to that in which they would otherwise have lost phase coherence. As a result their instantaneous precession frequencies differ less from the mean than would be the case with no motion, and narrowing of the resonance line is observed.

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 <sup>&</sup>lt;sup>1</sup> I. J. Lowe and R. E. Norberg, Phys. Rev. 107, 46 (1957).
 <sup>2</sup> J. H. Van Vleck, Phys. Rev. 74, 1168 (1948).

<sup>&</sup>lt;sup>8</sup> F. Bloch, preceding paper, Phys. Rev. 111, 841 (1958)

<sup>&</sup>lt;sup>4</sup> Bloembergen, Purcell, and Pound, Phys. Rev. 73, 679 (1948).

Another example is the use of macroscopic motion of the sample to average out magnetic field inhomogeneities.<sup>5</sup> Instead, however, of providing physical motion of the nuclei in question, an alternative procedure is to cause the local fields themselves to vary rapidly by perturbing the sources of these fields.<sup>6</sup> The double resonance experiments of Bloom and Shoolery,<sup>7</sup> Anderson,8 and Herzog and Hahn9 are examples of this approach.

In the present experiment, the 100% abundant isotopes,  $Na^{23}$  and  $F^{19}$ , in NaF are chosen as nuclei A and B respectively. Thus the  $F^{19}$  nuclei produce the local fields we wish to average out. A strong resonant perturbation is applied in the form of an oscillating rf magnetic field of amplitude  $2H_2$  and (angular) frequency  $\omega_2 = 2\pi\nu_2$  near the Larmor frequency  $\omega_0'$  of these nuclei, causing rapid transitions between the Zeeman levels. When this resonance perturbation exceeds the F-F dipole interaction, the local fields produced by the F<sup>19</sup> dipoles then fluctuate at a frequency  $\gamma' H_2/2\pi$ , where  $\gamma'$  is the gyromagnetic ratio of the F<sup>19</sup> nuclei. If this frequency can be made appreciably greater than the Na-F interaction frequency itself, the averaging may be expected to become effective. A more quantitative expression of this restriction is considered in reference 3. The nuclear resonance of the Na<sup>23</sup> nuclei is simultaneously observed by exploring the region about their Larmor frequency  $\omega_0$  in the same external magnetic field  $H_0$  with a weak rf magnetic field of amplitude  $2H_1$  and frequency  $\omega_1 = 2\pi \nu_1$ . The second moment,  $\langle \Delta_1^2 \rangle_{AV} = \langle (\omega_1 - \omega_0)^2 \rangle_{AV} = 4\pi^2 \langle (\Delta \nu_1)^2 \rangle_{AV}$ , of the observed Na<sup>23</sup> resonance is then determined as a function of the strong rf field  $H_2$  and of the deviation from resonance  $\Delta_2 = \omega_2 - \omega_0'$  of the F<sup>19</sup> nuclei.

As already mentioned, the second moment is expressible as

$$\langle [(\Delta \nu)^2]_{\mathrm{Na(total)}} \rangle_{\mathrm{Av}} = \langle [(\Delta \nu)^2]_{\mathrm{Na-Na}} \rangle_{\mathrm{Av}} + \langle [(\Delta \nu)^2]_{\mathrm{Na-F}} \rangle_{\mathrm{Av}}$$

and the last term is the only portion of the total that can be altered with this technique. The desirability of as large an effect as possible led to the particular choice of sample. Using Van Vleck's formulas, one computes  $\langle (\Delta \nu)^2 \rangle_{Av} = 9.66 \text{ (kc/sec)}^2$  for the total Na<sup>23</sup> second moment, with  $\langle [(\Delta \nu)^2]_{Na-Na} \rangle_{Av} = 1.89 \ (kc/sec)^2$ and  $\langle \lceil (\Delta \nu)^2 \rceil_{Na-F} \rangle_{Av} = 7.77 (kc/sec)^2$ . Thus the Na-F interaction is about 80% of the total. Neglected in this calculation are possible contributions to line broadening from the interaction of the nuclear electric quadrupole moment of Na<sup>23</sup> with electric field gradients produced by deviations from the ideally face-centered cubic symmetry of the NaF crystal structure. Experimental results justify this assumption. The present experiment

- <sup>7</sup> A. L. Bloom and J. N. Shoolery, Phys. Rev. 97, 1261 (1955). <sup>8</sup> W. A. Anderson, Phys. Rev. 102, 151 (1956).
- <sup>9</sup> B. Herzog and É. L. Hahn, Phys. Rev. 103, 148 (1956).

has dealt with powdered NaF only. Single crystals were not readily available; moreover, the demonstration of the effect did not require analysis of the dependence of the second moment upon the angle between crystal axes and the external magnetic field  $H_0$ .

It should be emphasized that not only is it necessary to satisfy the condition  $(\gamma' H_2/2\pi)^2 \gg \langle [(\Delta \nu)^2]_{\mathrm{Na}-\mathrm{F}} \rangle_{\mathrm{Av}}$ , as suggested before, but one must further require  $(\gamma' H_2/2\pi)^2 \gg \langle [(\Delta \nu)^2]_{\mathrm{F(total)}} \rangle_{\mathrm{Av}}$ . That is, the rf field must also be intense enough to "cover" the entire fluorine resonance line width, in order that all F<sup>19</sup> nuclei experience the coherent perturbation. A comparison of these conditions in the case of NaF reveals that the second is the more stringent:  $\langle [(\Delta \nu)^2]_{Na-F} \rangle_{Av} = 7.77$  $(\text{kc/sec})^2$ , whereas  $\langle [(\Delta \nu)^2]_{F(\text{total})} \rangle_{AV} = 99.21 (\text{kc/sec})^2$ . The maximum rf field used in the experiment was  $H_2 = 6.0$  gauss; therefore, with the known gyromagnetic ratio  $\gamma'$  of F<sup>19</sup>,  $(\gamma' H_2/2\pi)^2 = 576 \ (\text{kc/sec})^2$ . Thus the first inequality above is amply satisfied, whereas the second is not. We shall see that the experimental result of this failure to provide a sufficiently large rf field is to prevent the complete attainment of the averaging process. A further comparison of the above condition with that for saturation of the fluorine resonance will be enlightening. This latter condition may be expressed as  $(\gamma' H_2)^2 T_1 T_2 \cong 1$ , where  $T_1$  and  $T_2$  are respectively the thermal and phase-memory relaxation times of the fluorine nuclei. When these times were measured for the NaF sample used, the values obtained were  $T_1 \cong 1$ sec and  $T_2 \cong 2 \times 10^{-5}$  sec. Thus an rf field of approximately 10 milligauss is sufficient for saturating the resonance, but as seen above, this is entirely insufficient for the purpose of coherently perturbing all F<sup>19</sup> nuclei.

#### II. APPARATUS AND SAMPLE PREPARATION

Figure 1 shows a block diagram of the apparatus employed in the experiment.

The external polarizing magnetic field,  $H_0$ , about 4250 gauss, was provided by a regulated low-current electromagnet. This field was homogeneous over the 2-cm<sup>3</sup> sample volume to approximately 3 parts in 10<sup>5</sup>. A Pound-Knight type spectrometer<sup>10</sup> with some



FIG. 1. Block diagram of apparatus.

<sup>10</sup> R. V. Pound and W. D. Knight, Rev. Sci. Instr. 21, 219 (1950).

<sup>&</sup>lt;sup>6</sup> F. Bloch, Phys. Rev. **94**, 496 (1954); W. A. Anderson and J. T. Arnold, Phys. Rev. **94**, 498 (1954). <sup>6</sup> F. Bloch, Phys. Rev. **102**, 104 (1956), Sec. 7.

minor modifications was used to monitor the Na<sup>23</sup> resonance line shape near 4.8 Mc/sec. A potentiometertype chart recorder following a "lock-in" detector displayed the derivative of the absorption line shape.

Power at the resonant frequency of the F<sup>19</sup> nuclei was provided by a conventional "master oscillatorpower amplifier" capable of supplying 100 watts at 17 Mc/sec. This corresponded to a rotating component of the rf magnetic field of 8 gauss. The rf power was link-coupled to a low-impedance transmission line and matched to the transmitting coil with a capacitive divider, part of which resonated the coil. Voltages of the order of 1000 volts were developed across this resonant element, which was contained in the same shield box or "head" as the Pound-Knight coil. Therefore, it became important to decouple these two coils to prevent pickup of the high voltage and subsequent blocking of the Pound-Knight oscillator-detector. This was accomplished in part, of course, by the circumstance of two different operating frequencies, but more thoroughly by an orthogonal mounting of the coils and the placing of a 17-Mc/sec rejection filter between the Pound-Knight oscillator tube and its coil. The use of ceramic and Teflon coil forms and the introduction of forced air cooling were necessary owing to the temperature rise associated with the power dissipation in the "head" and sample.

The measurement, to about 5%, of the strong rf magnetic field was accomplished with a carefully calibrated nonresonant pickup coil and rf voltmeter. A graphical plot of the proportionality between this pickup voltage and the voltage across the resonant circuit was used to determine the rf field once the sample was inserted and this volume no longer was accessible to the pickup coil. This proportionality was rechecked several times during the course of the experiment.

The sample was prepared by compressing approximately 5 grams of reagent-grade powdered sodium fluoride in a steel cylinder. Nearly 50 000 lb/in.<sup>2</sup> was transmitted through a steel piston to the powder, and the resulting "slug" of material was  $\frac{1}{2}$  inch in diameter,  $\frac{3}{4}$  inch long, and chalky in appearance, and its density was about 2.0 g/cm<sup>3</sup>. Strains in the sample resulting from this preparation procedure will contribute an undesirable effect to the magnetic resonance. Such strains lead to a departure from the cubic symmetry ideally associated with the small crystallites comprising the powder.

The resulting electric field gradients are not the same throughout the sample, and therefore yield a range of interactions with the Na<sup>23</sup> electric quadrupole moments. To first order, the Na<sup>23</sup>  $(I = \frac{3}{2})$  resonance line is divided into an unshifted line at the Na<sup>23</sup> Larmor frequency and a set of flanking lines spread over a large frequency range determined by the range of quadrupole interactions. The central component contains 0.4 of the

original intensity, the remaining 0.6 being lost in the unresolvable side lines. The result is a worsening of the signal-to-noise ratio and also the introduction of a further complication. Van Vleck's expression for the second moment of the line shape has to be modified<sup>11</sup> because of the unequal spacing of the energy levels brought about by the quadrupole interaction.

In an effort to circumvent this difficulty, an attempt was made to anneal out strains in the sample. The slug was heated to about 800°C (mp 990°C) on a sheet of platinum and held there for several hours. It was then cooled at about 3°C/min to room temperature. The process was accompanied by a 25% decrease in volume, increasing the density to 2.5 g/cm<sup>3</sup>, within 90% of the single crystal value. In addition, the appearance of small crystallites, of dimensions perhaps 1 mm, were noted on the surface. The effectiveness of the procedure was demonstrated by observing that the intensity of the Na<sup>23</sup> resonance after annealing was approximately 2.3 times greater than before. This is to be compared with the factor 2.5 expected if the difference in intensity of the resonance before and after annealing were attributable entirely to the complete gathering-in of the side lines into the central resonance.

#### III. EXPERIMENTAL PROCEDURE AND RESULTS

The commonly used technique of varying the large laboratory field  $H_0$  for the purpose of sweeping through the resonance signal could not conveniently be used in this experiment. It was essential to preserve the simultaneous condition of resonance for both nuclear species. Thus the use of a field sweep for monitoring the Na<sup>23</sup> resonance would have necessitated a synchronous sweep of the frequency applied to the F<sup>19</sup> nuclei, with the attendant problems of ganging and padding several variable capacitors.

One concession to the maintenance of the two resonance conditions was made, however. The sinusoidal modulation of the magnetic field had the inevitable consequence of varying the exact position of the F19 resonance condition, since  $\omega_2 \equiv \omega_F$  was kept fixed during a run. Ideally, then, one would prefer to substitute frequency modulation of the Pound-Knight oscillator. However, great difficulty with the inevitable small attendant amplitude modulation ensues, since it behaves as a pseudonuclear signal. For this reason, the field modulation technique was retained. This effect was minimized by using the smallest modulation amplitude that led to an acceptable signal-to-noise ratio for the Na<sup>23</sup> signal. A peak-to-peak modulation amplitude of 1.5 gauss was employed. This corresponded to 6 kc/sec for the F<sup>19</sup> nuclei, and since the F<sup>19</sup> resonance had a full width at half-maximum height of about 20 kc/sec, the effect mentioned was indeed kept small, the variation occurring entirely within the F19 line width. Thus the monitoring of the Na<sup>23</sup> resonance line shape

<sup>11</sup> K. Kambe and J. F. Ollom, J. Phys. Soc. Japan 11, 50 (1956).



D C FIELD FIXED AT 4250 GAUSS

FIG. 2. Schematic definition of experimental procedure.

was accomplished by slowly varying the frequency of the Pound-Knight oscillator through the region of resonance and well on either side to provide a reference base line. Frequency markers were inserted on the chart by manual operation of a switch which supplied a pulse of voltage to the recorder input.

Measurements of the Na<sup>23</sup> thermal relaxation time in the NaF sample were made and yielded a value of about 5 sec for  $T_1$ . Such a large value dictates the use of a very small rf field  $H_1$  to prevent saturation and the use of a very slow frequency sweep to ensure sufficient time for readjustment of the equilibrium. Therefore, the level of oscillation was maintained at 30 millivolts across the resonant circuit, corresponding to an rf field of 2 milligauss; and a frequency sweep rate of about 500 cycles per second per minute was used. With an output time constant of 25 sec and the above-mentioned parameters, a signal-to-noise ratio of about 25 to 1 was obtained.

The modulation period, about 0.01 sec, is clearly much greater than  $T_2$ . It is, however, much shorter than  $T_1$ , and one may properly consider effects such as Halbach has calculated.<sup>12</sup> In the present situation, where one is constrained to use very small values of  $H_1$ , the possible distortion of the recorded absorption could not exceed 2%, an unobservable effect.

The procedure is represented schematically in Fig. 2. In the first set of runs (I) the quantity  $\Delta_2 \equiv \Delta_F$  was fixed at zero. This condition was determined by adjusting the ratio of  $\omega_F$  to  $\omega_{Na}$  to the value 3.55655, as computed from Walchli's tables.<sup>13</sup> This ratio was not a priori the precise one to use for the solid NaF sample, since the possible existence of small chemical shifts could not be excluded. These might alter the ratio by a maximum amount of the order of  $\pm 0.00050$ . It will be seen, however, that an experimental justification exists for the assumption that these shifts are very small.

The Na<sup>23</sup> resonance was then recorded several times for each of six different amplitudes of  $H_2$ , and the second moments  $\langle \Delta_1^2 \rangle_{Av} = 4\pi^2 \langle (\Delta \nu)^2 \rangle_{Av}$  computed in a

manner to be described. As discussed in the accompanying paper,<sup>3</sup> the total second moment is an invariant under the conditions of the experiment. The reason that an apparent change is observed to take place is the result of the limitation placed upon measurement by the signal-to-noise ratio in combination with the detailed behavior of the change in the line shape that occurs. Bloch shows<sup>3</sup> that this change consists of the pushing out from the central line of two symmetrically placed sidebands as the amplitude of  $H_2$  is increased from zero. As  $H_2$  increases, the sidebands move further out and their intensity decreases very rapidly; so rapidly, in fact, that by the time they could have been resolved from the center line, they are already "lost" in the noise. Thus, for a time, while they are still within the line width of the central line, the measurement of the second moment includes their contribution and  $\langle \Delta_1^2 \rangle_{AV}$  is constant. As soon, however, as they are lost in the noise, the cutoff which must be made in the mechanical computation of  $\langle \Delta_1^2 \rangle_{AV}$  excludes them from contributing. As a result the measured second moment of the remaining central line is observed to decrease, the difference between that value and the single resonance value being carried away by the sidebands. The theory referred to above<sup>3</sup> is valid only for sufficiently large values of  $H_2$ . Therefore, the manner in which  $\langle \Delta_1^2 \rangle_{Av}$  decreases in these runs will not be described by such a theory.

In the second set of runs (II),  $H_2$  was set at the largest convenient value so that some comparison with the theory could be made. For reasons of stability this value was limited to  $H_2=6.0$  gauss. This value was sufficiently large to insure that the sideband position was held well outside of the  $\omega_{Na}$  sweep range. The quantity  $\Delta_2 \equiv \Delta_F = \gamma' H_0 - \omega_2 \equiv \gamma_F H_0 - \omega_F$  was then adjusted to eight different values and several measurements of  $\langle \Delta_1^2 \rangle_{Av}$  were made under each of these conditions. The variation in  $\Delta_{\rm F}$  was accomplished by shifting  $H_0$  by the desired amount,  $\omega_{\rm F}$  always remaining fixed. Thus, the position of the Na<sup>23</sup> resonance was also shifted a small amount, but the range of sweep of  $\omega_{Na}$ was adjusted to take this into consideration. Equations (102) and (103) of the preceding paper then predict that the variation of the second moment with the parameter  $\Delta_{\mathbf{F}}$  should be described by a modified Van



FIG. 3. First derivative of Na absorption under single-resonance conditions,  $H_2=0$  (dashed line); and under double-resonance conditions,  $H_2=4.5$  gauss (solid line).  $\Delta_{\rm F}=0$ . Modulation equaled 1.5 gauss, peak-to-peak.

K. Halbach, Helv. Phys. Acta 27, 259 (1954).
 H. E. Walchli, Oak Ridge National Laboratory Report ORNL-1469 (unpublished).

Vleck equation

$$\langle (\Delta_1)_{6^2} \rangle_{Av} = \langle (\Delta_1^2)_{Na-Na} \rangle_{Av} + \langle (\Delta_1^2)_{Na-F} \rangle_{Av} \cos^2\theta, \quad (1)$$
  
where  
$$\cos^2\theta = (\Delta_F)^2 [(\gamma_F H_2)^2 + (\Delta_F)^2]^{-1}.$$

In the region about  $\Delta_{\rm F} = 0$ , both positive and negative values of  $\Delta_{\rm F}$  were chosen. The essential symmetry in  $\langle \Delta_1^2 \rangle_{\rm AV}$  that was observed, independent of the sign of  $\Delta_{\rm F}$ , lent confidence to the earlier-mentioned assumption of negligible chemical shifts. That is, the frequency ratio initially chosen seemed indeed to be correct for the NaF sample used. This ratio could not have been more accurately chosen in advance unless apparatus had been available with which the F<sup>19</sup> resonant frequency could have been measured for the same value of  $H_0$  as the Na<sup>23</sup> frequency. After a discussion of the technique used to evaluate  $\langle \Delta_1^2 \rangle_{\rm AV}$ , the results of these runs will be examined in detail.

Figure 3 is a replotting of recorder chart tracings of two derivative curves for the purposes of illustration only. The signal-to-noise ratio has been accidentally improved by the selection of only a limited number of points in the replotting. The total length of signal, plus baseline included in those specimens actually measured, was twice that depicted in Fig. 2. The calculation of  $\langle \Delta_1^2 \rangle_{Av}$  was not made from the derivative curve data, as has been suggested,<sup>14</sup> but rather from the integrated line shape. The reason for this procedure was twofold. First, it was of interest to examine the intensity of the resonance under various conditions of the experiment. This required that the derivative curve be point-wise numerically integrated so that the resonance line shape could be plotted. Then this resulting curve had to be integrated to determine the intensity. Second, a correction had to be point-wise incorporated into the numerical computation of  $\langle \Delta_1^2 \rangle_{Av}$ 



FIG. 4. Run No. I. Na resonance absorption curves obtained by integrating experimental derivative curves. The second rf field oscillates with a frequency equal to the F precessional frequency, i.e.,  $\Delta_F = 0$ .



FIG. 5. Run No. II. Na resonance absorption curves obtained by integrating experimental derivative curves. The second rf field is kept fixed in magnitude, i.e.,  $H_2=6$  gauss.

to eliminate the distortion of the derivative curve by the long output time constant. This correction would have involved the slope of the recorded derivative curve if it had been used directly in computing  $\langle \Delta_1^2 \rangle_{Av}$ . If the integrated line shape is used to compute  $\langle \Delta_1^2 \rangle_{Av}$ , however, the correction involves only the value of the ordinate of the recorded curve, and a considerable simplification in the handling of the data results. The major problem involved in the processing of the data arose from small displacements of the base line during a run, which the long time constant and the inherent inertia in the recorder prevented from complete selfaveraging. Upon integration, this led to resonance curves that did not return to the initially chosen base line. The asymmetry made them unsuited to a measurement of  $\langle \Delta_1^2 \rangle_{Av}$ , until a revised base line was drawn through the tails of the curve to give a best fit, as determined by eye and a flexible ruler. This method, while not excluding the possibility of some subjective bias, was the only consistent and reasonably simple one available.

It is apparent from Fig. 3 that the reduction of the second moment under conditions of double irradiation does not manifest itself in a marked decrease in width between peaks of the derivative curve. The most striking characteristics of the recorded curves are loss of intensity in the wings and the improvement of the signal-to-noise ratio by virtue of an increased peak derivative amplitude. The latter results from an increased slope at the inflection point of the integrated line shape.

From the value of  $\langle \Delta_1^2 \rangle_{\text{Av}}$  thus determined, was subtracted the correction term arising from the use of a finite modulation amplitude.<sup>15</sup> For a modulation amplitude of 1.5 gauss peak-to-peak, this correction amounted to 0.18 (kc/sec)<sup>2</sup>.

Figures 4 and 5 represent some of the integrated line

<sup>15</sup> E. R. Andrew, Phys. Rev. 91, 425 (1953).

<sup>&</sup>lt;sup>14</sup> G. E. Pake and E. M. Purcell, Phys. Rev. 74, 1184 (1948).



FIG. 6. Measured second moment of the Na resonance vs the amplitude of the second rf field for Run No. I.

shapes of runs I and II respectively. The original integrating process provides some smoothing of the data, since finite intervals must be used in the process. In addition, in the preparation of the sample curves for the figures, some further smoothing results from the choice of points for plotting. Except for slight differences in normalization, the curve of largest peak amplitude in Fig. 4 represents the same experimental situation as the curve of the largest peak amplitude in Fig. 5. The curve of least peak amplitude in Fig. 4 represents the single-resonance data. Except in the extreme tails, it is fitted very closely by a Gaussian line shape. The doubleresonance curves all have greatly reduced intensity in the wings, thus providing the reduction in second moment observed quantitatively. These double-resonance line shapes deviate markedly from a Gaussian function. All curves have the same total intensity within the accuracy of measurement possible, about 10%, and no trend in these measurements was observable. Thus, no nuclear Overhauser effect was observed.

Reference should be made to a possible resonance frequency shift<sup>16</sup> of the double-resonance lines. Because the two frequencies differed by a ratio appreciably greater than unity, and since the rf fields were of the oscillating and not rotating type, the expected shift would be no more than 0.8 cycle/second. The center cross-over point of a noisy, 5-kc/sec-wide derivative curve could clearly not be located to an accuracy approaching the above value.

Figure 6 summarizes the data from the series I runs. The single-resonance data lie about 0.4  $(kc/sec)^2$  above the Van Vleck value. This may be caused by two conditions. The annealing process may have been insufficient to gather the quadrupole side lines completely into the central line. Residual intensity in the wings from this source would enhance the single-resonance second moment. Also the known inhomogeneity in  $H_0$  will add to the line width. If a simple type of additive dependence can be assumed for the functions involved, the value of this spread in field over the sample is easily

shown to contribute very nearly the observed enhancement. The theory<sup>3</sup> shows that the satellites are well separated only if  $\gamma' H_2$  is large compared to the line width. This condition is not met for  $H_2=0.75$  gauss, and therefore all the intensity is still contained in the center main line. Thus, the second moment in Fig. 6 shows no deviation from the single-resonance value, since all components of the resonance are included in the measurement. For  $H_2=1.5$  gauss, however, an appreciable fraction of the sideband is outside of the center line and lost in the noise. It thus is prohibited from contributing to  $\langle \Delta_1^2 \rangle_{AV}$  by reason of the computational technique previously described. The degree of inadequacy of the maximum rf used is evident in the same drawing, since in the limit of very large values of  $H_2$ , one expects to approach asymptotically the dashed line representing the like-neighbor interaction. While this limit is evidently not reached, its approach is clearly indicated by the plot of Fig. 6.

The best indication of the accuracy involved is the spread in the data themselves. Each point represents an independent resonance recording, but at least two and usually four runs were made sequentially to preserve closely the experimental conditions. This met with only limited success, since the time required for a single run was in excess of 70 minutes. The solid curve through the experimental points is illustrative only, and has no simple theoretical significance.

Figure 7 summarizes the data from the series II runs. The single-resonance runs are again shown for comparison. The lower points at  $\Delta_{\rm F}/2\pi=0$  correspond exactly to the extreme right-hand points in Fig. 6. Measurements at  $\Delta_{\rm F}/2\pi = -10$  kc/sec and -20 kc/sec were also made. As mentioned earlier, they were situated nearly as mirror reflections in the Y axis of the points shown in the figure for the corresponding positive values of  $\Delta_{\rm F}/2\pi$ . At  $\Delta_{\rm F}/2\pi=240$  kc/sec, the conditions were essentially again those of single resonance, since the frequency ratio then deviated by an amount equivalent to about ten line widths from



FIG. 7. Measured second moment of the Na resonance vs the deviation of the oscillation frequency of the second rf field from the F precessional frequency for Run No. II.

<sup>&</sup>lt;sup>16</sup> N. F. Ramsey, Phys. Rev. 100, 1191 (1955).

that required for simultaneous resonance. There is an apparent tendency for these points to lie even somewhat higher than the single-resonance points obtained for  $H_2=0$ . However, the decreased stability resulting from operating at an elevated temperature causes a greater spread in the data. Thus it is probably unwarranted to attach much significance to this difference. The solid curve has been calculated from Eq. (1). To improve the fit with the data, the observed excess of  $0.42 \ (kc/sec)^2$  in the second moment of the single resonance line has been added. The agreement is satisfying in the region beyond  $\Delta_{\rm F}/2\pi = 20$  kc/sec. The discrepancy near the ordinate axis, i.e., for small values of  $\Delta_{\mathbf{F}}$ , is not unexpected, since  $H_2$  alone was insufficient to satisfy completely the conditions of the theory.<sup>3</sup> The theoretical curve in Fig. 7 is therefore unreliable in this region.

Further study in this field may profitably deal with the use of single-crystal samples and the added information that comes from analysis of the dependence of the second moment on the angle between the crystal axes

and  $H_0$ . It is also of interest to compare  $T_1$  of the type-A spin system in the single- and double-resonance cases in a crystal containing two magnetic ingredients, A and B. When spin diffusion<sup>17</sup> is the predominant thermal relaxation mechanism for the A spin system, then the averaging out of local fields by the simultaneous double resonance of the B system could be expected to increase the spin diffusion rate of the A spin system. A resulting shortening of the  $T_1$  of the A system would then occur. Experiments of the latter type are being carried out in this laboratory for the case of a single crystal of LiF where the longer thermal relaxation time of Li<sup>7</sup> simplifies the experimental details.

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<sup>17</sup> N. Bloembergen, Physica 15, 386 (1949).

## PHYSICAL REVIEW

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# **Two-Particle Interactions in Deformed Nuclei\***

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Effects of two-particle interactions in deformed nuclei are considered. The deformed nucleus is assumed to be replaced by a set of particles in a cylindrically symmetric harmonic oscillator potential, while the twoparticle interaction is assumed to be of  $\delta$ -function type. Fairly extensive calculations are performed in diagonalizing the interaction energies which exist between particles lying in degenerate levels in the abovementioned potential. The characteristic energies and the explicit form of the eigenfunctions are tabulated. In particular it is noticed that the seniority number is a good quantum number for this sort of interaction. Several important features of the results are discussed, including the matrix elements of the E2 transitions.

## 1. INTRODUCTION

HE unified model of the nucleus developed by Bohr, Mottelson,<sup>1,2</sup> and others<sup>3</sup> is known to be quite powerful in explaining many low-energy nuclear phenomena. The basic idea underlying this model is explained in detail in the literature, especially in Chap. V of reference 3.

The success of this model was most remarkable for

rotational motions in that the concept of a stable deformation resulted directly in the rotational spectra. The low-lying levels of nuclei away from closed shells can be qualitatively interpreted in this way.

More quantitative investigations taking into account the single-particle motions in such deformed nuclei were performed by Nilsson,<sup>4</sup> Moszkowski,<sup>5</sup> and Gottfried,<sup>6</sup> and their results have been found to explain quite well a number of experimental data.4-7

The problem concerning the moment of inertia is more involved, because here the correlation of the particle motions is relevant. It has been investigated theoreti-

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