

Table I. Summary of experimental results

Run	Irrad. time (h)	$\frac{\Delta a_0}{a} \times 10^4$	$\Delta g_0$ (nΩ cm)	Annealing range (°K)	$\frac{\Delta a/a}{\Delta a_0/a}$ (%)	$\frac{\Delta g}{\Delta g_0}$ (%)	$\frac{3 \Delta a/a}{\Delta g_0}$ (mΩ <sup>-1</sup> cm <sup>-1</sup> )
I	46.0	2.71 ± 0.08	124 ± 2 <sup>a</sup>	4-60	38 ± 5	31 ± 2	8.0 ± 2.0
				60-300	46 ± 5	51 ± 2	5.9 ± 1.0
				300-1,200	16 ± 5	18 ± 1	5.8 ± 1.6
II	86.3	3.75 ± 0.15	190 ± 2 <sup>b</sup>	4-60	36 ± 5	29 ± 1.5	7.4 ± 1.5
				60-300	48 ± 5	54 ± 1.5	5.2 ± 0.6
				300-1,200	16 ± 5	17 ± 0.5	5.6 ± 1.6

<sup>a</sup>From measurements in the same irradiation position

<sup>b</sup>Measured simultaneously with a different sample at the same irradiation position

duced than the resistivity per defect per atom compared with the single-defect values.

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<sup>1</sup>W. Bauer, A. Seeger, and A. Sosin, Phys. Letters **24A**, 195 (1967).

<sup>2</sup>R. O. Simmons and R. W. Balluffi, Phys. Rev. **109**, 1142 (1958).

<sup>3</sup>R. W. Balluffi and R. O. Simmons, J. Appl. Phys. **31**, 2284 (1960).

<sup>4</sup>R. Doll, H. Meissner, N. Riehl, W. Schilling, and F. Schmeissner, Z. Angew. Phys. **17**, 321 (1964).

<sup>5</sup>H. Meissner, W. Schilling, and H. Wenzl, EuroNuclear **2**, 277 (1965).

<sup>6</sup>A. Sepp, U. Himmeler, H. Peisl, H. Wenzl, and W. Waidelich, Phil. Mag. **14**, 131 (1966).

<sup>7</sup>U. Himmeler, H. Peisl, A. Sepp, W. Waidelich, and H. Wenzl, Z. Angew. Phys. **23**, 8 (1967).

<sup>8</sup>A. Seeger, E. Mann, and R. v. Jan, J. Phys. Chem. Solids **23**, 639 (1962).

<sup>9</sup>G. W. Iseler, H. I. Dawson, A. S. Mehner, and I. W. Kaufmann, Phys. Rev. **146**, 468 (1966).

<sup>10</sup>M. Rühle, Phys. Status Solidi **19**, 263 (1967).

## TEMPERATURE DEPENDENCE OF ULTRASONIC PARAMAGNETIC RESONANCE IN MgO:Fe<sup>2+</sup> †

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The technique of ultrasonic paramagnetic resonance (UPR) provides a particularly direct means of investigating electron-spin-phonon interactions.<sup>1,2</sup> Because most UPR measurements have been made at 10 GHz, at which frequency the background acoustic attenuation rises rapidly above 4°K, they have been restricted to liquid-helium temperatures. At lower microwave frequencies, at which the acoustic attenuation permits measurement over a wider range of temperature, the pulse techniques<sup>1,2</sup> most often used are not sensitive enough to observe UPR in most paramagnetic ions.

In this paper we report measurements of

the UPR of Fe<sup>2+</sup> ions in MgO at 1.1 GHz over a temperature range of 4.2 to 30°K. Measurements were made on the temperature dependence of the amplitude of both the absorption and dispersion signals and on the temperature dependence of the linewidth. To our knowledge these are the first reported measurements of either UPR absorption or dispersion over an extended temperature range. The measurements were made using a sensitive continuous wave (cw) ultrasonic technique which involves exciting a mechanical resonance in the sample.<sup>3</sup> Ultrasonic absorption decreases the Q of the mechanical resonance line while ultra-

sonic dispersion shifts the mechanical resonance frequency. The measurements were made on a crystal of MgO containing approximately 20 ppm of  $\text{Fe}^{++}$  ions. The sample, 0.510 in.  $\times$  0.560 in.  $\times$  0.280 in. in length, was prepared with faces flat to 0.1 wavelength of sodium light and parallel to 2 sec of arc. Only the central section, 0.250 in. in diameter, was used for the experiment. The sample faces were gold plated for ground electrodes. CdS transducers were deposited on top of the gold. All of the measurements were made with longitudinal acoustic waves propagating in the  $\langle 100 \rangle$  direction.

The effective spin of  $\text{Fe}^{++}$  in MgO is 1. Because it exhibits much less strain broadening than the  $\Delta m = \pm 1$  transition and is equally allowed by the "quadrupolar" transition probabilities,<sup>4</sup> measurements were made on the  $\Delta m = \pm 2$  transition. For this transition one can derive an attenuation coefficient  $\sigma_2$  which describes the amplitude attenuation, due to a resonant interaction with electron spins, of a longitudinal acoustic wave propagating along a cube axis of the MgO crystal<sup>4</sup>:

$$\sigma_2 = \frac{9n\pi\omega G_{11}^2}{32\rho\hbar v^3} g(\omega) \sin^4\theta, \quad (1)$$

where  $n$  is the population difference per unit volume between the spin levels involved (+1 and -1),  $\omega$  is the angular frequency of the acoustic wave,  $\rho$  is the crystal density,  $v$  is the acoustic wave velocity,  $\theta$  is the angle between the direction of acoustic wave propagation and the external magnetic field,  $g(\omega)$  is the UPR line-shape function, and  $G_{11}$  is the magnetoelastic coupling constant.

Associated with the resonant change in the attenuation of the acoustic wave is a resonant change in the phase velocity<sup>5</sup> which may be written as

$$\frac{\Delta v}{v} = \left(\frac{v}{\omega}\right) \frac{\omega_0 - \omega}{\delta} \sigma_2, \quad (2)$$

where  $\delta$  is a measure of the linewidth and  $\omega_0$  is the frequency corresponding to an absorption maximum. Internal strains in the MgO crystal lead to an asymmetric  $\Delta m = \pm 2$  line shape and determine the linewidth at low enough temperatures.<sup>6</sup> It is expected that at high enough temperatures the linewidth will be determined by lifetime broadening due to a short  $T_1$ .

The  $\Delta m = \pm 2$  absorption linewidth was measured as a function of temperature at 1.1 GHz.

The width between peak-to-peak points of the derivative signal is difficult to measure accurately for this asymmetric line. A more sensitive parameter<sup>6</sup> is the distance between the  $\frac{1}{4}$ - and  $\frac{3}{4}$ -height points of the maximum peak-to-peak deflection of the derivative signal. This width is presented in Fig. 1. With the exception of the 10-13°K temperature region the width shows the expected behavior. Above 16°K the line broadens steadily until it is lost in noise above 26°K. Below 16°K the linewidth is approximately constant with the exception of the region just below 13°K. It appears that some unexpected mechanism broadens the line between 10 and 13°K. It is believed that the linewidth below 16°K and outside the 10-13°K range is determined by a random distribution of strains. From a measurement of the linewidth in this region, the strain distribution in the MgO crystal was estimated to be about  $10^{-5}$ , in good agreement with the value obtained from epr measurements on  $\text{Mn}^{++}$  and  $\text{Fe}^{+++}$  in MgO.<sup>7</sup> The rapid variation in linewidth between 10 and 13°K was the same for all runs. This broadening is not understood at the moment.

The linewidth at temperatures above 16°K is believed to be determined partially by lifetime broadening. The spin-lattice relaxation time,  $T_1$ , at these temperatures is expected to be due to either a Raman or Orbach process. The data indicate an Orbach process. The linewidth at 16°K is determined mainly by internal strains while the linewidth at the highest temperature is determined mainly by  $T_1$  broadening. In order to obtain the contribution of

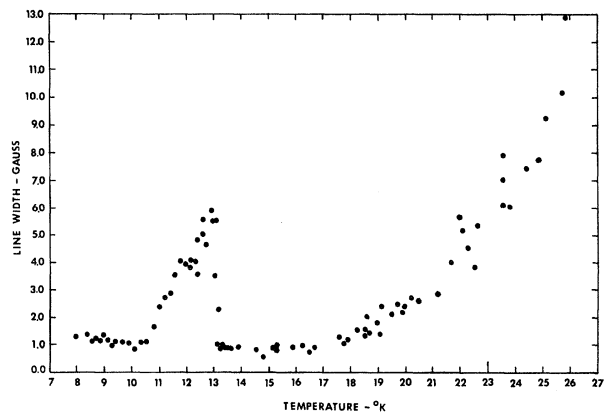


FIG. 1. UPR  $\Delta m = \pm 2$  absorption signal linewidth versus temperature in  $\text{MgO}:\text{Fe}^{2+}$  at 1.1 GHz. Longitudinal acoustic waves are propagating along  $[100]$  direction  $\perp H$ .

$T_1$  to the linewidth, the data above 16°K are handled in the following way: A corrected width is determined by subtracting the measured width at 16°K from the measured width at the higher temperatures. Figure 2 is a plot of the corrected linewidth in frequency units versus inverse temperature. The conversion to frequency units is made by using  $\Delta\omega = \gamma\Delta H$  with  $\gamma = 5.8 \times 10^7 \text{ sec}^{-1} \text{ G}^{-1}$ .  $\Delta H$  is the linewidth of Fig. 1 minus the linewidth at 16°K. The solid line in Fig. 2 represents the equation

$$\Delta\omega = A \exp(-\Delta/T), \quad (3)$$

with  $A = 2.6 \times 10^{11} \text{ sec}^{-1}$  and  $\Delta = 155^\circ\text{K}$ . The interpretation given to Fig. 2 is that over the temperature range 16-26°K the spin-lattice relaxation proceeds via an Orbach process involving an excited state which is 155°K above the ground state. The solid line in Fig. 2 was obtained by fitting the data by eye. Various lines drawn in this manner give a value for the excited level of  $155 \pm 5^\circ\text{K}$ ; however, in order to take into account the uncertainty concerning the exact correction to apply to the linewidth (the subtraction of the 16°K width), it is felt that a more realistic estimate of the error is  $\pm 10^\circ\text{K}$ .

Recent epr measurements<sup>8</sup> of the  $\Delta m = \pm 2$  transition of  $\text{Fe}^{++}$  in MgO show an Orbach contribution to  $T_1$ . The epr work was carried

out at X band and in the temperature range 1.2-15°K. The Orbach contribution was found to be

$$(T_1)^{-1} = B \exp(-\sigma/kT), \quad (4)$$

with  $B = 6.5 \times 10^{10} \text{ sec}^{-1}$  and  $\delta = 133^\circ\text{K}$ . The magnitude of  $T_1$  is difficult to obtain from our results because, although  $T_1$  is inversely proportional to  $\Delta\omega$ , the proportionality constant is not precisely known for the  $\Delta m = \pm 2$  asymmetric line. If it is assumed that  $\Delta\omega \approx (T_1)^{-1}$ , our results are in fair agreement with the results of Ref. 8. It should be noted that one experiment (epr) was a study of the  $\Delta m = \pm 1$  transition between 1.2 and 15°K with X-band microwaves. The second experiment (UPR) was a study of the  $\Delta m = \pm 2$  transition between 16 and 26°K with 1.1-GHz ultrasound. Infrared-absorption measurements<sup>9</sup> made on Fe-doped MgO reveal an excited level at 151°K which is attributed to  $\text{Fe}^{++}$ . The result presented here for the excited level is in good agreement with the results of the infrared measurements.

Measurements of the UPR absorption signal amplitude showed the signal increasing rapidly with temperature between 4.2 and 9°K. We attribute this to a saturation of the signal at the lower temperatures. With the exception of the 10-13°K temperature region, the temperature dependence of the signal is explained quite well by Eq. (1).  $n$  is given by

$$n = n_0 / (1 + 2UT_1), \quad (5)$$

where  $n_0$  is the equilibrium population difference and  $U$  is the ultrasonic transition probability for the spin levels involved. The anomalous broadening between 10 and 13°K causes the signal amplitude to depart from the expected value in this region.

Measurements were also made of the small changes in phase velocity (dispersion signal) associated with the spin-phonon interaction. The dispersion signal amplitude was found to be related to the absorption signal amplitude, as predicted by Eq (2), at the higher temperatures. The behavior of the dispersion signal at low temperatures was quite different from the behavior of the absorption signal. Figure 3 shows the dispersion signal amplitude plotted versus inverse temperature. The data were taken at a frequency of 1.1 GHz. The angle between the direction of propagation of the ultrasonic wave and the magnetic field was 90°.

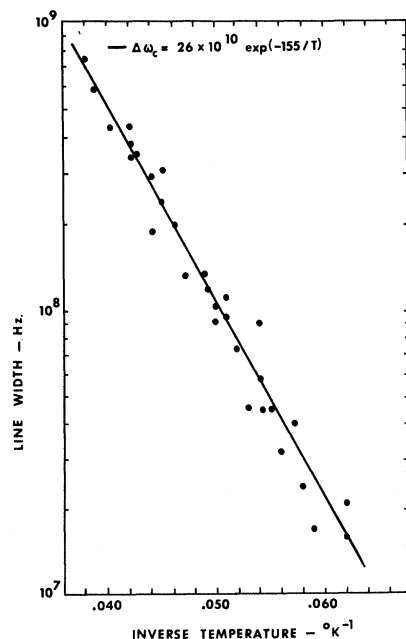


FIG. 2. Corrected absorption linewidth versus inverse temperature. Conditions as in Fig. 1.

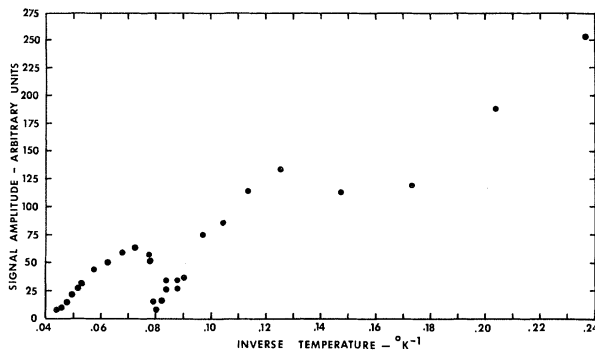


FIG. 3. UPR  $\Delta m = \pm 2$  dispersion signal amplitude versus temperature. Conditions as in Fig. 1.

The dispersion signal also shows anomalous behavior between 10 and 13°K. The dispersion signal shows no sign of saturation at low temperatures. This agrees with saturation measurements on UPR absorption and dispersion at 4.2°K.<sup>10</sup> No theory has yet been given to describe the behavior of the UPR dispersion signal under conditions of saturation. The experimental behavior of the UPR absorption and dispersion signals under conditions of saturation is similar to the behavior of the epr absorption and dispersion signals. In the epr case also, the absorption signal saturates much

easier than the dispersion signal.<sup>11</sup> The fact that the temperature dependence of the absorption and dispersion signals is qualitatively different suggests that it may be best to use the dispersion mode when searching for new signals.

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<sup>1</sup>E. B. Tucker, in *Physical Acoustics*, edited by W. P. Mason, (Academic Press, Inc., New York, 1966), Vol. 4A.

<sup>2</sup>R. Guerneur, J. Joffrin, A. Levelut, and J. Penne, *Phys. Letters* **13**, 107 (1964).

<sup>3</sup>D. I. Bolef, J. de Klerk, and R. B. Gosser, *Rev. Sci. Instr.* **33**, 631 (1962).

<sup>4</sup>R. D. Mattuck and M. W. P. Strandberg, *Phys. Rev.* **119**, 1204 (1960).

<sup>5</sup>E. H. Jacobsen and K. W. H. Stevens, *Phys. Rev.* **129**, 2036 (1963).

<sup>6</sup>D. H. McMahon, *Phys. Rev.* **134**, A128 (1964).

<sup>7</sup>E. R. Feher, *Phys. Rev.* **136**, A145 (1964).

<sup>8</sup>R. L. Hartman, E. L. Wilkinson, and J. G. Castle, Jr., *Bull. Am. Phys. Soc.* **12**, 642 (1967).

<sup>9</sup>J. Y. Wong and A. L. Schawlow, *Bull. Am. Phys. Soc.* **12**, 655 (1967).

<sup>10</sup>R. Guerneur, J. Joffrin, A. Levelut, and J. Penne, *Phys. Letters* **15**, 203 (1965).

<sup>11</sup>A. M. Portis, *Phys. Rev.* **91**, 1071 (1953).

### PROTON SINGLE-PARTICLE STATES ABOVE $Z = 82$ , AS OBSERVED WITH THE REACTION $^{208}\text{Pb}(^3\text{He}, d)^{209}\text{Bi}$ AT $E_{^3\text{He}} = 51 \text{ MeV}$ \*

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The location and identification of the proton single-particle states above  $Z = 82$  is a problem of basic interest<sup>1,2</sup> and the subject of recent<sup>3-5</sup> discussion and investigation. This note presents the experimental angular distributions of the six strong transitions observed to bound states of  $^{209}\text{Bi}$  in an investigation of the reaction  $^{208}\text{Pb}(^3\text{He}, d)^{209}\text{Bi}$  at  $E_{^3\text{He}} = 51.26 \text{ MeV}$ . An analysis of these data with local, zero-range distorted-wave (DW) calculations<sup>6</sup> yields a positive identification of the first five of these levels with the  $1h_{9/2}$ ,  $2f_{7/2}$ ,  $1i_{13/2}$ ,  $2f_{5/2}$ , and  $3p_{3/2}$  single-particle states, respectively.

The experiment was performed with the Oak Ridge Isochronous Cyclotron and the associat-

ed broad-range spectrograph facility. The target was a 0.47-mg/cm<sup>2</sup> lead foil of isotopic constitution 95%  $^{208}\text{Pb}$ , 1.2%  $^{207}\text{Pb}$ , and 3.8%  $^{206}\text{Pb}$ .<sup>7</sup> The experimental energy resolution (full width at half-maximum) for the  $^{209}\text{Bi}$  groups was 60 keV. The major target contaminants were  $^{12}\text{C}$  and  $^{16}\text{O}$ ; the  $(^3\text{He}, d)$  reaction products from these nuclei obscured the particle groups of one or two levels of  $^{209}\text{Bi}$  at several angles. The absolute cross sections of the  $(^3\text{He}, d)$  transitions to the levels of  $^{209}\text{Bi}$  were established by reference to the elastic yields of  $\text{Pb}(^3\text{He}, ^3\text{He})\text{Pb}$  at  $\theta_L = 14^\circ, 16^\circ, 18^\circ, \text{ and } 25^\circ$ . The cross sections for this elastic scattering were assumed to be those predicted in an optical-model cal-