

the 804-keV gamma-ray, a conversion coefficient is obtained for this radiation of 20 to 30 percent. This value of the conversion coefficient would require the 804-keV gamma-radiation to be  $M4$ , but considerations of parity and angular momentum<sup>5</sup> in this transition between two even-even nuclei indicate the radiation must be electric.

If, in fact, the internal conversion coefficient for the 804-keV gamma-ray is about 5 percent and the soft component is  $K$ -radiation, the above intensity measurements indicate that most of the  $K$ -shell ionization is due to processes other than internal conversion, for example, ionization by alpha-particles.<sup>6</sup>

In an attempt to resolve these difficulties a search was made for a possible time association between the two components. A spectrum of coincidences was plotted in the region of 80 keV, with one channel set on the high energy component only. No significant coincidences to the 77-keV component were observed with a resolving time of 0.3 microsecond, and it was concluded that less than 1 percent of the soft radiation is in coincidence with the 804-keV component. This was taken as evidence that, even if part of the soft radiation is nuclear in origin, a prompt cascade de-excitation process does not exist.

<sup>1</sup> K. Siegbahn and S. H. E. Slatis, *Nature* **159**, 471 (1947).

<sup>2</sup> Zajac, Broda, and Feather, *Proc. Phys. Soc. (London)* **60**, 501 (1948).

<sup>3</sup> Grace, Allen, West, and Halban, *Proc. Phys. Soc. (London)* **64**, 493 (1951).

<sup>4</sup> D. Alburger and G. Friedlander, *Phys. Rev.* **81**, 523 (1951).

<sup>5</sup> S. De Benedetti and G. H. Minton, *Phys. Rev.* **85**, 944 (1952).

<sup>6</sup> Since the completion of the above work, W. C. Barber and R. H. Helm have reported similar results [*Phys. Rev.* **86**, 275 (1952)]. They find the ratio of the number of soft rays to the number of 800-keV gamma-rays to be  $0.134 \pm 0.025$  to 1, and suggest that only about one-third or one-half of the  $K$ -shell ionization is due to internal conversion, the remaining ionization being a direct result of alpha-emission.

## Wall Energy of Ferroelectric Domains

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THE present paper will give a simple tentative explanation of the behavior of small ferroelectric particles reported in a previous letter.<sup>1</sup>

The stable domain configuration of an isolated and non-conducting ferroelectric crystal corresponds to a minimum of the sum of wall energy and volume energy. The arrangement of the domains which has to be expected for a  $\text{KH}_2\text{PO}_4$ -crystal is shown in Fig. 1.<sup>2</sup> Assuming a cube-shaped crystal with edges  $D$ , consisting of  $n$  domains and imbedded in a medium with the dielectric constant  $\epsilon$ , the depolarization energy is<sup>3</sup>

$$W_{\text{dep}} = 1.7P^2D^3/n\epsilon. \quad (1)$$

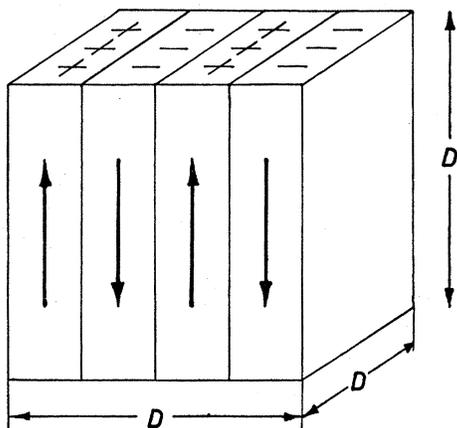


FIG. 1. Expected arrangement of the domains for a  $\text{KH}_2\text{PO}_4$  crystal.

For the surface energy density of the domain walls, we may put  $\sigma = \alpha P^2$ . Hence,

$$W_{\text{wall}} = (n-1)\alpha P^2 D^2. \quad (2)$$

These energies are now introduced into the well-known Mueller interaction theory.<sup>4</sup> According to Mueller the free energy density of the shorted (single domain) crystal is

$$A = (T - T_C)P^2/2C + \frac{1}{4}\xi'P^4 + \dots, \quad (3)$$

where  $T_C$  is the Curie temperature ( $= 123^\circ\text{K}$ ),  $C$  is the constant of the Curie-Weiss law ( $= 260^\circ\text{C}$ ),<sup>2</sup> and the coefficients  $\xi', \dots$  are assumed to be independent of temperature.

Introducing  $W_{\text{dep}}$  and  $W_{\text{wall}}$  into Eq. (3), the total free energy of the free energy of the crystal described above turns out to be

$$Aa^3 = a^3[P^2\{T - T_C + 3.4C/n\epsilon + 2(n-1)\alpha C/D\}/2C + \xi'P^4/4 + \dots]. \quad (4)$$

Comparing this expression with (3), we find that the Curie temperature is lowered by

$$\Delta T_C = -\{3.4C/n\epsilon + 2(n-1)\alpha C/D\}. \quad (5)$$

The stable domain number  $n$  corresponds to a minimum of  $|\Delta T_C|$ . Hence, for given  $\epsilon$  and  $D$ , the relation between  $\Delta T_C$  and  $\alpha$  is unique.

For small particles the second term in (5) becomes important. The measurement of the Curie temperature of small isolated particles allows, therefore, the evaluation of the wall energy coefficient  $\alpha$ . The previously reported investigations on colloidal  $\text{KH}_2\text{PO}_4$  have now been completed by dielectric measurements with an improved sample technique.<sup>5</sup> A lowering of the Curie temperature with decreasing  $\epsilon$  and decreasing  $D$  has been observed.

For  $\epsilon \sim 5$  and  $D \geq 4000\text{Å}$  the Curie point was found to be above the temperature of liquid nitrogen (i.e.,  $|\Delta T_C| < 46^\circ\text{C}$ ). From this an upper limit for  $\alpha$  can be deduced:

$$\alpha < 26 \times 10^{-8} \text{ cm.}$$

On the other hand, for  $\epsilon \sim 5$  and  $D \leq 1500\text{Å}$  the Curie point is below the temperature of liquid nitrogen (i.e.,  $|\Delta T_C| > 46^\circ\text{C}$ ), indicating a lower limit

$$\alpha > 9 \times 10^{-8} \text{ cm.}$$

These limits are consistent with the assumption of classical dipole-dipole interaction together with a domain wall thickness of a few lattice constants.<sup>6</sup>

The value of the spontaneous polarization being  $2.5 \times 10^{-6}$  coulomb/cm<sup>2</sup>, the limits for the surface energy density of a domain wall are

$$20 < \sigma < 58 \text{ erg/cm}^2.$$

The author is indebted to Professor P. Scherrer for illuminating discussions.

<sup>1</sup> W. Käzig and M. Peter, *Phys. Rev.* **85**, 940 (1952).

<sup>2</sup> See, e.g., Baumgartner, Jona, and Käzig, *Ergeb. Exakt. Naturwiss.* **23**, 235 (1950).

<sup>3</sup> C. Kittel, *Revs. Modern Phys.* **21**, 541 (1949).

<sup>4</sup> See, e.g., W. G. Cady, *Piezoelectricity* (McGraw-Hill Book Company, Inc., New York, 1946), p. 580.

<sup>5</sup> The dielectric measurements reported in the first paper (see reference 1) are not conclusive, as the colloid had undergone a change from being pressed into disks.

<sup>6</sup> C. Kittel, *Phys. Rev.* **82**, 965 (1951).

## Anomalous Magnetic Resonance Absorption of $\text{Cu}^{++}$

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IN magnetic resonance absorption of salts containing  $\text{Cu}^{++}$ , no structure is observed in the resonance lines except for hyperfine structure. The reason is that the ion is in a  $2D$  state and

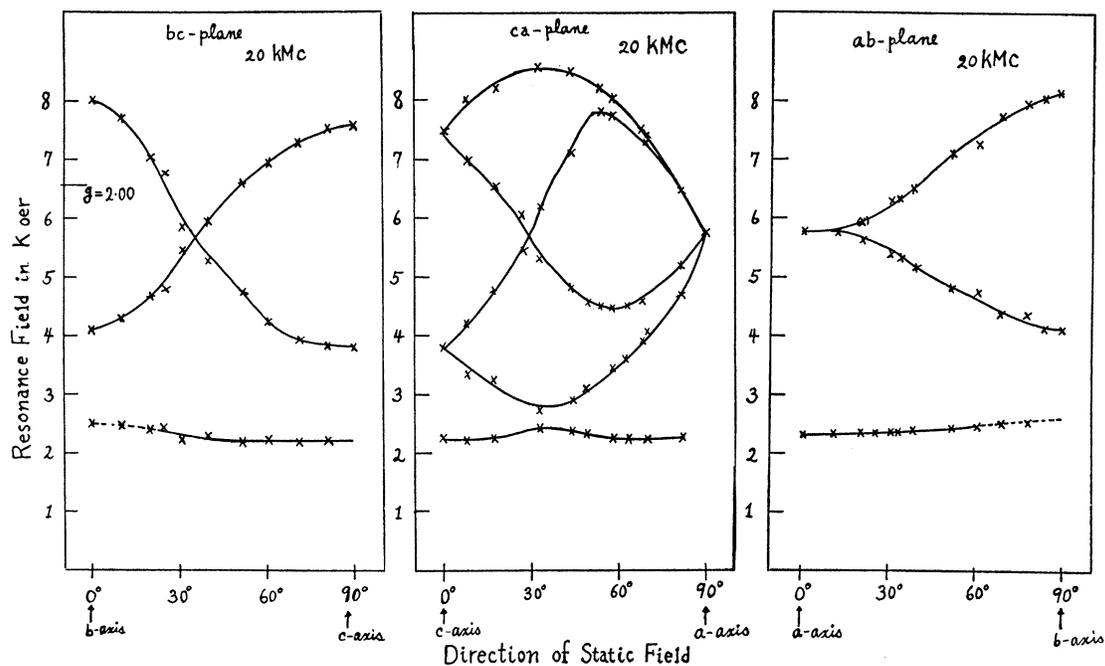


FIG. 1. Peak resonance field strengths observed at 20 kMc when the static magnetic field is in the  $bc$ ,  $ca$ , and  $ab$  planes, respectively. The  $a$ ,  $b$ , and  $c$  axes are magnetic axes.

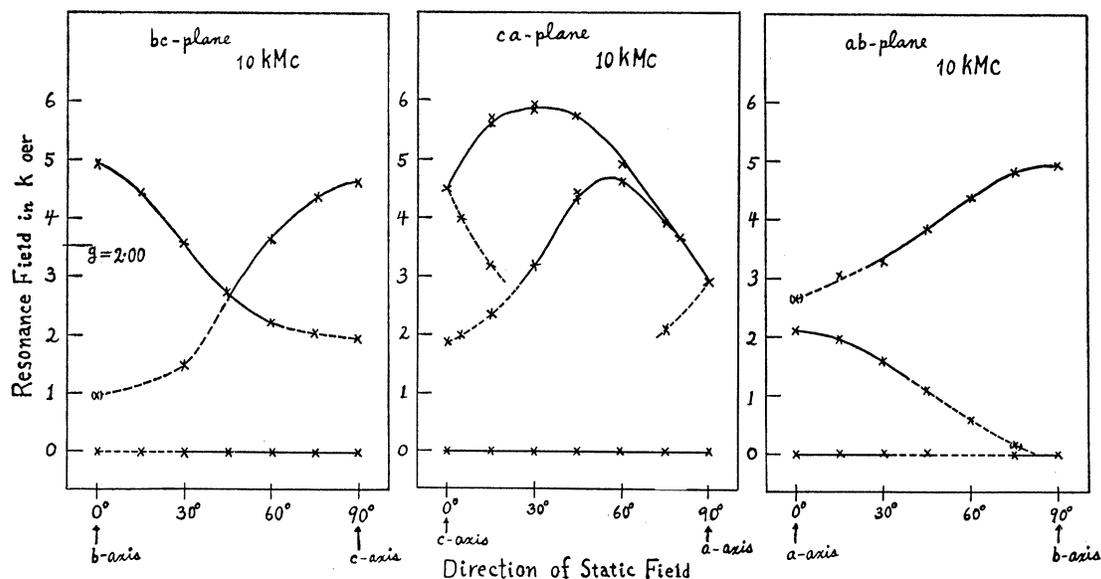


FIG. 2. Peak resonance field strengths observed at 10 kMc when the static magnetic field is in the  $bc$ ,  $ca$ , and  $ab$  planes, respectively.

the ground state in crystalline fields is split only by an external magnetic field, the upper levels being far above the ground state. The  $g$ -value is affected by the upper level and varies with the orientation of the static magnetic field. It ranges from 2.05 to 2.20 as in  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ ,<sup>1-3</sup>  $\text{CuK}_2\text{Cl}_4 \cdot 2\text{H}_2\text{O}$ , etc.<sup>4,5</sup>

Recently, however, Lancaster and Gordy<sup>6</sup> have pointed out anomalous resonance absorption in copper acetate powder. We have measured the absorption in single crystals of this salt at 10 and 20 kMc. The composition of this salt is  $\text{Cu}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$ , the number of water molecules being determined by chemical analysis.

The field strengths at the observed peaks are shown in Figs. 1 and 2. The  $a$ ,  $b$ , and  $c$  axes in the figures are magnetic ones, and not axes of crystal forms. The data observed at 20 kMc are shown in Fig. 1, and the corresponding data at 10 kMc, observed by Hayashi and Ôno in our laboratory, are shown in Fig. 2. Up to five peaks are observed, whose  $g$ -values are distributed in a wide range. At 10 kMc, a peak seems to exist at about zero magnetic field, showing that the salt has a zero-field splitting of about 10 kMc. The peaks in the  $a$ -direction in the second and third parts of Fig. 2 do not coincide; this point is under examination now. Dotted lines show that the absorption is

weak. The microwave magnetic field is always perpendicular to the static field.

Such anomalous absorptions cannot be explained by a single ion in the  $^2D$  state in any allowable crystalline field. The first possibility for the origin of this anomaly may be that ions are not in the  $^2D$  state in this case of very strong crystalline fields. The second possibility is that the absorption is not effected by a single  $\text{Cu}^{++}$  ion, but by a molecule containing  $\text{Cu}^{++}$  and other atoms. The third one is that a number of  $\text{Cu}^{++}$  ions are located at small distances from each other so that their electron spins mutually interact strongly. We note that the absorption of  $\text{Mn}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}$  shows a single peak with  $g=2.0$ .

The authors' thanks are due to Assistant Professors K. Kambe and E. Ishiguro for their discussions. A more detailed report will appear in *J. Phys. Soc. Japan*.

<sup>1</sup> D. M. S. Bagguley and J. H. E. Griffiths, *Proc. Roy. Soc. (London)* **201**, 366 (1950).

<sup>2</sup> R. D. Arnold and A. F. Kip, *Phys. Rev.* **75**, 1199 (1949).

<sup>3</sup> J. Wheatley and D. Halliday, *Phys. Rev.* **75**, 1412 (1949).

<sup>4</sup> Itoh, Fujimoto, and Ibamoto, *Phys. Rev.* **83**, 852 (1951).

<sup>5</sup> H. Kumagai *et al.* (to be published).

<sup>6</sup> F. W. Lancaster and W. Gordy, *J. Chem. Phys.* **19**, 1181 (1951).

## Electron-Hole Recombination in Germanium

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POWER rectifiers have been described that consist of thin wafers of high purity germanium whose opposite faces are heavily doped with donor and acceptor impurities.<sup>1</sup> Since the current flow depends largely upon the generation and recombination of holes and electrons within the high purity region, this geometry is well adapted to the study of the recombination process. A two-body collision mechanism leads to a rate that is proportional to the square of the carrier concentration at high levels of injection where holes and electrons are present in nearly equal numbers. Measurements show, however, that the rate varies linearly with concentration over a wide range of concentration and temperature.<sup>2</sup> These observations can be accounted for by assuming that recombination takes place largely through the agency of recombination centers distributed throughout the germanium.

A steady-state recombination rate given by<sup>3</sup>

$$R = (np - n_i^2) / [t_p(n + n_0) + t_n(p + p_0)], \quad (1)$$

results from a simple model in which the centers give rise to an energy level lying in the forbidden band. The electron and hole concentrations are given by  $n$  and  $p$ , their product under equilibrium conditions being  $n_i^2$ . The lifetime for electrons when the centers are completely empty is given by  $t_n$ ;  $t_p$  is the hole lifetime with all centers occupied by electrons. The recombination centers lie at an energy level defined by  $n_0$  and  $p_0$  which are the equilibrium electron and hole concentrations in a sample whose Fermi level coincides with the position of the recombination centers.

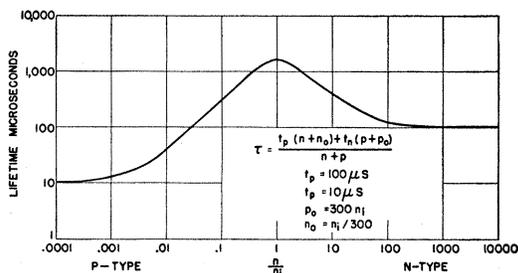


FIG. 1. Variation of lifetime with impurity content.

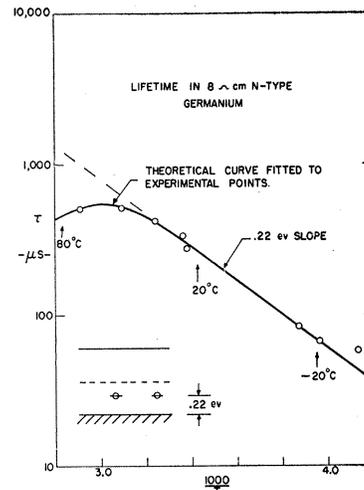


FIG. 2. Variation of lifetime with temperature.

In the derivation of Eq. (1), it is assumed that the rate of capture of electrons is proportional to the number of empty centers and to the number of free electrons. The rate at which electrons are emitted by these centers is proportional to the number of occupied centers. Analogous statements apply to the capture and emission of holes.

It is readily seen that Eq. (1) can account for the linear relation between carrier concentration and recombination rate mentioned in the first paragraph. With high level injection,  $n$  and  $p$  become equal and much greater than  $n_i$ ,  $n_0$ , or  $p_0$  so that  $R \approx n / (t_n + t_p)$ . Rectifier characteristics calculated on the basis of Eq. (1) can be fitted to the data in a very satisfactory manner.<sup>4</sup> The lifetime for holes and electrons at high level injection ( $t_n + t_p$ ) is usually found to be approximately 100  $\mu\text{sec}$ .

The equilibrium lifetime which results from Eq. (1) is

$$\tau \equiv \lim_{\delta n \rightarrow 0} (\delta n / \delta R) = [t_p(n + n_0) + t_n(p + p_0)] / (n + p). \quad (2)$$

The variation of  $\tau$  with impurity content is illustrated in Fig. 1 for a choice of parameters which gives reasonable agreement with lifetimes observed in samples of germanium at room temperature.

Since  $n_0$  and  $p_0$  vary exponentially with  $1/T$  while  $t_n$  and  $t_p$  should be relatively insensitive to temperature, the lifetime should assume its limiting values  $t_n$  and  $t_p$  for  $p$ - and  $n$ -type samples at low temperatures. At higher temperatures, the lifetime increases with temperature as long as the sample remains extrinsic and then decreases again in the intrinsic range. Measurements of lifetime illustrating the variation with temperature in the upper two temperature ranges are shown in Fig. 2. The slope indicated on the graph gives a tentative value of 0.22 electron volt for the position of the recombination centers above the valence band or below the conduction band.

<sup>1</sup> R. N. Hall and W. C. Dunlap, Jr., *Phys. Rev.* **80**, 467 (1950).

<sup>2</sup> R. N. Hall, *Phys. Rev.* **83**, 228 (1951).

<sup>3</sup> This expression was presented in the text of reference 2, using somewhat different notation. Shockley has derived it as a special case of a more general treatment which has been submitted for publication.

<sup>4</sup> A detailed treatment of the germanium power rectifier will be published shortly.

## An Experimental Test of the Shell Model

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IN a recent letter, Bethe and Butler<sup>1</sup> proposed an experiment to give direct information on the accuracy of the shell model of nuclear structure in ascribing definite orbital angular momen-