Scintillation Response of Anthracene to Low-Energy Protons and Helium Ions*

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Photomultiplier pulse amplitudes produced by light from anthracene crystals bombarded by protons and helium ions from 25 to 375 kev were measured. Both response curves (pulse height *vs* energy) are nonlinear, but that for protons bends away from the energy axis while that for helium ions bends towards it. Both curves extrapolate smoothly to the origin, indicating an absence of any nonscintillating surface layer more than about 10 kev thick. Present knowledge of stopping cross sections (dE/dx) for anthracene is inadequate to permit an unambiguous determination of the specific fluorescence (dL/dx) from the observed pulse amplitudes. However, some qualitative conclusions can be drawn concerning Birks' theory of the fluorescent process.

I. INTRODUCTION

THE emission of light from certain crystals bombarded by charged particles has been of considerable interest.[1–4] Organic crystals, such as anthracene, can be used to count individual slow ions if the response to such particles is known. Also, the light output, measured by the pulse amplitude produced in a photomultiplier tube, may give information about the luminescent process itself. The response of sodium iodide for slow heavy particles has been reported,[5,6] but all previous experiments with anthracene and other organic crystals have been done by slowing down particles with initial energies of several Mev. Because of straggling and uncertain range-energy relations, the low-energy response was poorly determined. In this experiment, ions accelerated by the Nebraska Cockcroft-Walton generator to accurately known energies between 25 and 375 kev were used to bombard anthracene crystals and the response curve has thus been extended below 100 kev.

II. EXPERIMENTAL METHOD

Figure 1 shows the experimental arrangement. The crystal chamber A was joined directly to the accelerator vacuum system. Protons, negative hydrogen ions, and singly charged helium ions were drawn from an rf ion source containing argon contaminated with a few percent of hydrogen and helium; this was necessary to reduce the intensity of the desired ion beam to a few hundred particles per second so that they could be counted individually. The energies of these ions were known within 2% from the generator voltage which was measured with a potential divider and potentiometer.[7] A magnetic analyzer was used to deflect the desired ions through about 40°, down the axis of the tube leading to the crystal chamber. They then passed through a $\frac{1}{16}$-in. hole in the center of the spherical aluminum mirror M, used to increase the efficiency of collection of light from the crystal at X.

Light from the crystal was transmitted to the cathode of the photomultiplier P through the Lucite window L, shaped to fit the curved surface of the phototube. Optical contact between L and P was made with Nujol mineral oil. In some runs, anthracene crystals about 1.0 cm×1.0 cm×0.4 cm were used; optical contact between these crystals and the window was also made with Nujol oil. Some thin (about 0.5 mm) crystals were also used. These were cemented to small glass blocks which were sealed to the window with oil. The aperture of the mirror and the diameter of the window were $1\frac{1}{4}$ in.; the effective solid angle for light from the crystal was about 3π.

The photomultiplier tube used was a special 17-stage tube developed by Schaetti,[8] and characterized by high gain, exceptionally low noise, and very high

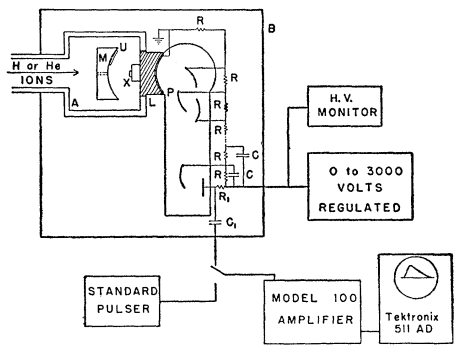

FIG. 1. Block diagram of apparatus.

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[1] J. B. Birks, *Scintillation Counters* (McGraw-Hill Book Company, Inc., New York, 1953), and S. C. Curran, *Luminescence and the Scintillation Counter* (Academic Press, Inc., New York, 1953) contain extensive bibliographies.

[2] G. T. Wright, Phys. Rev. **96**, 569 (1954); J. Sci. Instr. **31**, 462 (1954); Phys. Rev. **91**, 1282 (1953).

[3] G. T. Wright and G. F. J. Garlick, Brit. J. Appl. Phys. **5**, 13 (1954).

[4] W. H. Robinson and W. Jentschke, Phys. Rev. **95**, 1412 (1954).

[5] S. K. Allison and H. Casson, Phys. Rev. **90**, 880 (1953).

[6] F. S. Eby and W. K. Jentschke, Phys. Rev. **96**, 911 (1954).

[7] Cook, Jones, and Jorgensen, Phys. Rev. **91**, 1417 (1953).

[8] N. Schaetti and W. Baumgartner. Z, angew. Math. u Phys. I. 268 (1950); Helv. Phys. Acta **23**, 524 and 869 (1950); Le Vide, No. 34–35, 1041 (1951).

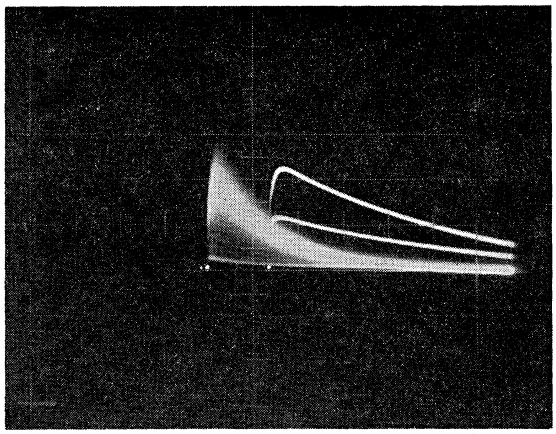

FIG. 2. Pulse distribution for 275-kev protons on anthracene. The sharp lines are reference pulses.

vacuum. The tube produces fewer than 2000 noise pulses per second, and these are almost certainly caused by single electrons from the cathode. The cathode quantum efficiency is about that for an RCA 5819. Since most of the pulses from ions were well above the single-electron pulses for incident energies of 75 kev or more, it would seem that the 5819 or similar tube could be used for efficient counting of these particles, the noise being biased off.

Dynode voltages were supplied by a resistance divider, the last three dynodes being by-passed directly to the high-voltage supply. About 2200 volts was sufficient to produce at the anode a mean pulse height of over one millivolt for the least energetic particles. The mean pulse height varied with high voltage in agreement with the gain *vs* voltage curves supplied by the manufacturer for this particular tube. Throughout any day's run, the high voltage was continuously monitored by comparing a fraction of it to a $67\frac{1}{2}$-volt reference battery; the comparison was sufficiently sensitive to cause the mean pulse height to fluctuate less than 1% due to variation in tube voltage.

The pulses were amplified by a Los Alamos Model 100 amplifier with a rise time of about 0.6 microsecond and a clipping time of 8 microseconds. The pulses displayed on a Tektronix 511 AD oscilloscope were photographed with a Speed Graphic press camera giving a 4×5 inch negative which was used without further enlargement. A time exposure was taken sufficiently long to obtain more than three thousand pulses; this required less than one minute. Two standard pulses whose amplitudes bracketed the mean pulse amplitude of the ions were also placed on each photograph. The standard pulses were obtained by using a mercury relay (Western Electric 275A) to apply a measured voltage to an accurately-measured resistance attenuator; the relative sizes of the reference pulses were known to better than 2%. A sample photograph is shown in Fig. 2.

The mean pulse height was obtained by measuring the mean of the distribution of particle pulses, the height of the two standard pulses, interpolating, and converting to height in millivolts at the input of the amplifier. At the lower energies, where the pulse distribution becomes very broad, the determining of the "mean" pulse height by this method is questionable, since the judgment of the observer plays a significant role. However, above 100 kev, the mean amplitudes could be determined within 5 to 7%. The consistency between various measurements of the same photograph, and the consistency between various runs with different crystals was considerably better than this estimation, averaging about 3%.

III. EXPERIMENTAL RESULTS

Figure 3 summarizes the results of five runs with protons for four different anthracene crystals and of four runs with helium ions for three different crystals.† Each run normally gave a curve for both protons and helium ions with energies from 25 to 350 or 375 kev. Since the photomultiplier voltage could not be set exactly the same for each day's run, a single normalization point, chosen to be the smooth-curve pulse height for 250-kev protons, was used for each run. The bars shown on the experimental points are the average deviations from the mean of pulse height at a fixed

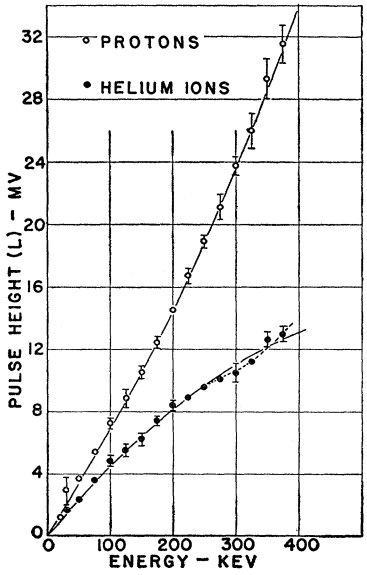

FIG. 3. Scintillation response of anthracene. ○ Protons. ● Helium ions. Ordinates are normalized pulse heights in millivolts at amplifier input. The solid curves are least-squares fitted to a quadratic form and are $L_p = 62E + 56E^2$ and $L_{He} = 50E - 42E^2$, with E in Mev.

† *Note added in proof.*—These results should be compared with the proton results of Fowler and Roos [Phys. Rev. **98**, 996, (1955), Fig. 2]. Their curve is somewhat more nonlinear, giving, for example, a ratio of 350-kev pulse height to 250-kev pulse height about 25% larger than obtained here. The cause of the difference is unknown.

energy after normalization; these average about 3% for both proton and helium ion curves. The various runs covered a time of several months, and the crystals were from two different batches supplied by the Harshaw Chemical Company.

In spite of the relatively high vapor pressure of anthracene, no contamination of the vacuum was observed. After several weeks *in vacuo*, the crystals developed a slight haze on the surface, but even this did not seem to affect the pulse-height distribution, which in every run taken, extrapolated smoothly to the origin. No fatigue or discoloration of the crystals was observed, but the integrated dose of heavy-particle bombardment was probably much too small to produce this effect.[1]

To be sure that the method was capable of producing correct results, a sodium iodide crystal was used and the response measured for protons and helium ions up to 250-kev energy. These data were in agreement with those obtained by Allison and Casson[5] for a similar experiment.

To facilitate comparisons with other experiments, the pulse height produced by the 624-kev internal conversion betas from Cs^{137} was measured, as well as that produced by the alphas from a natural uranium source having a mean energy of 4.3 Mev. On the scale used in Fig. 3, the 624-kev beta pulse height is 330 and the 4.3-Mev alpha pulse height is 230, both $\pm 7\%$. Using these data for normalization, the results of this experiment appear consistent with previous experiments at higher energies.[3,9]

Very rough estimates of the spread in pulse heights were made from the photographs. The percent resolution (full width at half-maximum divided by the mean pulse height) appeared consistent with the $E^{-0.7}$ variation found by Allison and Casson[5] for sodium iodide. For protons at 100 kev, the percent resolution was about 65%; it is estimated that at this energy the mean pulse size is about that which would be produced by pulses from the emission of four photoelectrons from the cathode.

IV. ANALYSIS AND DISCUSSION

Certain qualitative features of Fig. 3 are interesting. For protons, the curve of L (mean pulse height) *vs* energy is generally convex towards the energy axis, while that for helium ions is generally concave towards it. Thus the slope, dL/dE, for protons increases with energy, while that for helium ions decreases. Hence the light produced (dL) for a small energy loss (dE) in the crystal increases with the energy of the proton, but decreases with the energy of the helium ion. Since the maximum of the Bragg curve (specific ionization *vs* energy) for protons in anthracene is probably between 50 and 100 kev and for helium ions is probably between 250 and 400 kev, it is likely that, over most of the energy range observed in this experiment, the specific ionization for protons is decreasing while that for helium ions is increasing. Thus the observed curvature in the response curves is possibly correlated with the ionization density.

This is further substantiated by the possible inflection in the curve for helium ions, shown by the dotted line in Fig. 3. Not only does such an inflection point seem required by the data at higher energies,[6] but also the Bragg maximum for helium ions is near the energy of the possible inflection point in Fig. 3.

Since the response curves for both particles seem to continue in a very smooth way back to the origin, any surface effect tending to reduce the production of light is either very small or is a smooth function of the incident particle energy. An inactive surface layer thicker than about 10 kev seems definitely ruled out.

The response curves have generally been analyzed[1,4] by plotting the specific fluorescence, dL/dx, (x=range of particle) as a function of the differential energy loss with range, dE/dx. The slopes of the curves in Fig. 3 give values for dL/dE. If dE/dx is known, then dL/dx can be calculated. dE/dx can be found from the stopping cross sections, but unfortunately these have not been measured for the energy region below 500 kev for any solids other than metals[10] and ice.[11] By assuming the additivity of atomic stopping powers and assuming that the cross sections are independent of the physical state of the element, the stopping power of anthracene ($C_{14}H_{10}$) can be calculated from the work of Hirschfelder and Magee.[12] They computed, semiempirically, the stopping powers of several elements for energies as low as 5 kev, although the methods used may not be valid below several hundred kev. Using these values for dE/dx and combining them with the slopes from Fig. 3 (the dotted line was used for the helium ion curve) to get dL/dx, the results shown in Fig. 4 were obtained.

Figure 4 shows also the results of previous experiments taken from Robinson and Jentschke.[4] The linear response curve is obtained from electron data, not shown in this figure. The previous proton and alpha data are shown by the crosses. The alpha data show "saturation" of the specific fluorescence; that is, dL/dx becomes independent of dE/dx for large dE/dx. The proton dL/dx increases less than linearly with dE/dx, but there is no definite evidence of "saturation." (Although the ordinate of Fig. 4 is in "arbitrary units," normalization between various experiments is done in terms of the 624-kev beta pulse heights. Hence agreement of alpha and proton curves will indicate the degree of consistency between the experiments.)

Using the Hirschfelder-Magee dE/dx, the present

[9] Taylor, Jentschke, Remley, Eby, and Kruger, Phys. Rev. **84**, 1034 (1951).

[10] See a review article by S. K. Allison and S. D. Warshaw, Revs. Modern Phys. **25**, 779 (1953).

[11] W. A. Wenzel and W. Whaling, Phys. Rev. **87**, 499 (1952).

[12] J. Hirschfelder and J. L. Magee, Phys. Rev. **73**, 207 (1948).

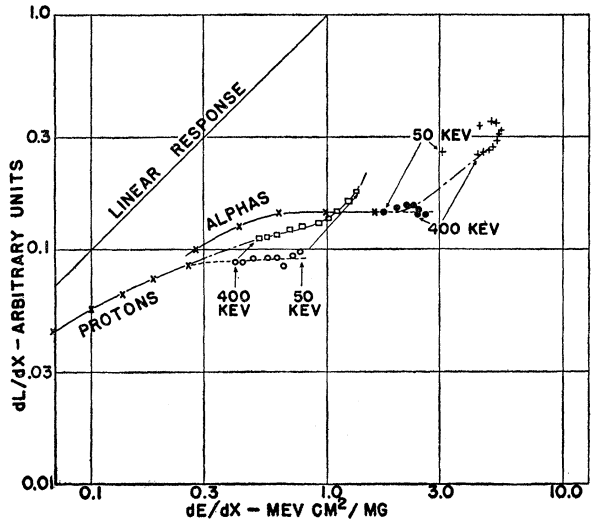

FIG. 4. Specific fluorescence *vs* differential energy loss for anthracene. Points × are taken from Robinson and Jentschke (reference 4). Points □ (protons) and + (helium ions) are derived using Hirschfelder-Magee stopping powers (reference 12). Points ○ (protons) and ● (helium ions) are derived from the "solids" stopping powers (see text). The "linear response" curve is determined by electron data, not shown here (reference 4).

proton data do lie on a smooth extension of the previous curve. However, two properties of this extension should be observed: (1) No evidence for saturation of specific fluorescence is seen at values for dE/dx much greater than values which produce saturation for alpha particles. (2) At very large dE/dx (low energies), dL/dx increases more than linearly with dE/dx. Such a situation is difficult to understand physically, although the theory of Chou[13] permits such a possibility. This was pointed out by Milton and Fraser,[14] who obtained a somewhat similar response curve for fission fragements in KI.

In a similar way, the present helium ion data also imply that (2) occurs for alpha particles, but only after the considerable region of saturation shown by the higher energy data. The up-turn of dL/dx must be rather abrupt, as shown by the dot-dash line in Fig. 4. Further, for the very high dE/dx obtained in this experiment, dL/dx appears to become a double-valued function of dE/dx. While this is possible physically, it is in a certain sense unsatisfactory.

It is possible that these results are produced by the use of incorrect values of dE/dx in anthracene. In an effort to obtain better values, the published data on the stopping powers of solids were reviewed,[10,11] and a set of stopping powers was derived on the following assumptions:

(1) Only experimental data for solids should be used (none of the data from experiments with gases was used).

(2) At each energy, the proton stopping cross section is a smooth function of atomic number.

(3) Atomic stopping cross sections for elements in a solid can be combined by addition to give the total cross section.

(4) The ratio of the alpha stopping cross section at $4E$ to the proton cross section at E is not four ($=Z^2$) but is that given by experiments on gold (the only solid for which comparisons have been made experimentally).

If the resulting cross sections are used with the slopes from Fig. 3, then the points labelled "Solids dE/dx" in Fig. 4 are obtained. These seem to fit with the high-energy data somewhat better than those obtained from the Hirschfelder-Magee dE/dx. Further, the saturation effect shows up consistently for both protons and alpha particles, and the present data for helium ions is in very good agreement with the observed saturation at lower values of dE/dx.

The saturation of specific fluorescence is expected on the basis of a simple theory proposed by Birks.[15] This gives

$$dL/dx=\phi(x/a_0)A\,(1+BdE/dx)^{-1}dE/dx, \qquad (1)$$

where A and B are constants, a_0 is a length characterizing the crystal, and ϕ is a function of x/a_0 which is 0.5 at zero range and which increases in a roughly exponential fashion to unity. From Fig. 4 and the previous experiments, it appears likely that in the present experiment for both protons and alpha particles, BdE/dx is sufficiently larger than unity that, approximately,

$$dL/dx=\phi A/B.$$

Since ϕ increases monotonically with energy, the present low-energy data should lie below the curves established by the high-energy experiments. Since, however, they actually lie above the previous data, or at most show saturation of specific fluorescence, ϕ cannot be varying greatly over this energy region. Hence a_0 must be rather small. Since the range of the most energetic protons used in this experiment is less than 6 microns in anthracene and that of the most energetic helium ions is less than 2 microns, it seems unlikely that a_0 can be as large as the 3 microns suggested by Birks.

A further conclusion on the basis of either value of dE/dx in Fig. 4 is that the constant B in Eq. (1) cannot be the same for both protons and alpha particles. (The constant A simply defines the scale for measuring pulse heights.) This substantiates the conclusion already reached by Robinson and Jentschke.[4]

It is, however, to be emphasized that an analysis of the present data according to Fig. 4 is not really possible until the stopping power, dE/dx, for protons and alphas in anthracene, or at least in some similar substance, is much better known than at present. It is most likely that neither set of points for the present data is correct, and Fig. 4 should not be taken too seriously. There is a great need for some accurate experimental measure-

[13] C. N. Chou, Phys. Rev. **87**, 903, 904 (1952).
[14] J. C. D. Milton and J. S. Fraser, Phys. Rev. **96**, 1508 (1954).
[15] See reference 1, p. 93.

ments of the stopping cross sections of solids at low energies.

V. ACKNOWLEDGMENTS

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Ferromagnetic Resonance in Two Nickel-Iron Ferrites

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Ferromagnetic resonance data are presented as a function of temperature from 4.2°K to 380°K in $(NiO)_{0.95}(FeO)_{0.05}Fe_2O_3$ and in $(NiO)_{0.75}(FeO)_{0.25}Fe_2O_3$. When the effects of eddy currents are corrected for, most of the energy dissipation in $(NiO)_{0.75}(FeO)_{0.25}Fe_2O_3$ as measured by the resonance line width is shown to be due to a relaxation mechanism associated with the presence of divalent iron. The line width shows a maximum at 160°K at our frequency of measurement (24 000 Mc/sec). A thermodynamical analysis of the loss mechanism is presented which assumes that a torque associated with part of the free energy relaxes. This correlates the line width quite satisfactorily with the temperature dependence observed in the magneto-crystalline anisotropy and the spectroscopic splitting (g) factor.

I. INTRODUCTION

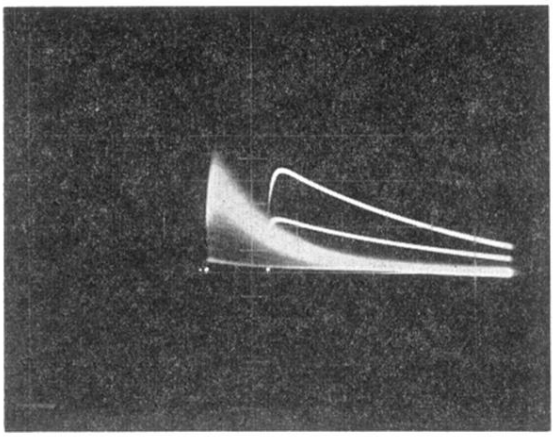
IN recent years, several studies of ferromagnetic resonance line width have been made in single crystals of ferrites.[1–5] It has, however, remained difficult to understand the mechanism of the losses in these materials. The purpose of the present paper is to report ferromagnetic resonance experiments on single crystals of two ferrite compositions which we feel throw some light on this mechanism. Quantitative chemical analysis for Ni and Fe together with the usual valences give $(NiO)_{0.95}(FeO)_{0.05}Fe_2O_3$ and $(NiO)_{0.75}(FeO)_{0.25}Fe_2O_3$ as the simplest formulas for the compositions on which we have worked. There is some possibility of small variations from these formulas in detail, but they are at least good approximations. The first of these compositions was grown by J. P. Remeika at Bell Laboratories using a slight variation on a method previously reported[6] which involves solutions of the constituent oxides in borax at high temperatures. The second composition was grown by Dr. G. W. Clark of the Linde Air Products Company by means of a flame fusion method.

Our data consist of: (1) resonant field as a function of crystal orientation, and (2) the width of the resonance line for both ferrites as a function of temperature from 4.2° to 380°K and as a function of crystal direction at several temperatures. Because they showed features of special interest, preliminary data on line width as a function of temperature in $(NiO)_{0.75}(FeO)_{0.25}Fe_2O_3$ have been reported previously.[7]

From our data we are able to deduce for each temperature the magnetocrystalline anisotropy constants K_1 and K_2, the general damping parameter λ, and the spectroscopic splitting factor g. The analysis used to deduce these parameters is simple and has been presented elsewhere.[1,2,8] We will therefore merely summarize results which we must use. The technique of this experiment will also be discussed only briefly, since it is very similar to previous work done by us on small oriented spheres in a microwave cavity.[2,3]

We present in more detail an elementary thermodynamical theory of the line-width data in terms of the idea that a large part of the losses in ferrites arise from relaxations associated with electronic rearrangements like that which occurs in the low-temperature transition in Fe_3O_4. We suggest that the maximum in the line width at 160°K which we see in the data on $(NiO)_{0.75}(FeO)_{0.25}Fe_2O_3$ (see Fig. 3) is due to such a relaxation associated with a rearrangement of the valence electrons on the iron ions as the magnetization moves. The suggestion that motion of these valence electrons is involved in losses has been made before by

[1] L. R. Bickford, Jr., Phys. Rev. **78**, 449 (1950). Technical Report XXIII Laboratory for Insulation Research Massachusetts Institute of Technology October, 1949 (unpublished).
[2] Yager, Galt, Merritt, and Wood, Phys. Rev. **80**, 744 (1950).
[3] Galt, Yager, Remeika, and Merritt, Phys. Rev. **81**, 470 (1951).
[4] D. W. Healy, Jr., Phys. Rev. **86**, 1009 (1952).
[5] T. Okamura and Y. Kojima, Phys. Rev. **86**, 1040 (1952); T. Okamura, Sci. Rept. Research Inst. Tôhoku Univ. **A6**, 89 (1954).
[6] Galt, Matthias, and Remeika, Phys. Rev. **78**, 391 (1949). Remeika grew the $(NiO)_{0.95}(FeO)_{0.05}Fe_2O_3$ used in the present experiments with sodium metaborate as a flux, rather than the sodium tetraborate used in this reference.
[7] Galt, Yager, and Merritt, Phys. Rev. **93**, 1119 (1954).
[8] C. Kittel, Phys. Rev. **73**, 155 (1948).



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