the left by about 0.03 ev, just the resolution of the analyzer. The general shape and half-width of the curve are in good agreement with theory. Even at room temperature the half-width of the normal-energy curve is almost twice the half-width of the total-energy curve.

VII. CONCLUSIONS

1. The spherical field-emission retarding-potential analyzer measures the total electron energy distribution.

2. The Fowler-Nordheim theory in the total-energy representation is further verified by this sensitive method of determining the source of field-emitted electrons.

3. The range of energy distribution half-widths available in field and thermionic emission is essentially the same.

4. The high temperature sensitivity of the energy distribution suggests its application for measuring the temperature of the emitting surface, which is difficult to do by other methods because of the minute size of the emitter.

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Effect of the Energy Gap on the Penetration Depth of Superconductors

A. L. Schawlow and G. E. Devlin Bell Telephone Laboratories, Murray Hill, New Jersey (Received August 25, 1958)

The dependence on temperature of the penetration depth of superconducting tin crystals has been measured by a new low-frequency (100 kc/sec) method. The sample serves as the core of a solenoid whose inductance changes with the penetration depth. The inductance controls the frequency of an oscillator which can be measured precisely. It is found that there are departures from the law $\lambda = \lambda_0 [1 - (T/T_c)^4]^{\frac{1}{2}}$ derived from the Gorter-Casimir two-fluid theory. The departures are shown to arise from an energy gap in the spectrum of electron excitations and are qualitatively like those predicted by Lewis' extension of the two-fluid model to include a gap. Throughout the temperature range from 1.8°K to 3.69°K the measured penetration depths agree well with the theory of Bardeen, Cooper, and Schrieffer.

I. INTRODUCTION

CEVERAL problems of current interest in super- \mathbf{J} conductivity involve the penetration of magnetic fields into the surface of superconductors. The very small magnitude of this penetration makes experiments on it difficult and often indirect. Nevertheless previous experiments have indicated ways in which superconductors depart from the Londons' phenomenological equations, and most of these departures have now been given a theoretical basis by the fundamental theory of Bardeen, Cooper, and Schrieffer.¹ Thus it seems likely now that the proper relation between current and field in a superconductor is nonlocal. One investigates this empirically by studying the magnetic properties of samples having at least one very small dimension, i.e., films, wires, or colloids,²⁻⁴ and comparing their properties with those deduced from the behavior of macroscopic superconductors. So one needs to determine separately the laws of penetration into large and small superconductors. The latter have the advantage of being obtainable as single crystals with controllable orientation and relatively unstrained surfaces.

It should be recalled that superconductors are almost perfectly diamagnetic. Only in a very thin surface laver, a few hundred angstroms deep, can there be any magnetic field. In this layer, currents flow which are just large enough to reduce the field inside the rest of the specimen to zero.

With a large superconductor, it is just about impossible to know its over-all size accurately enough for direct comparison with the penetration layer thickness, so one resorts to measuring the change in the penetration depth with temperature. Then if the law of temperature dependence is assumed known, there will be just one constant to fit to the experiments, and this can be the penetration depth at absolute zero.

It is possible to derive this law of temperature dependence in a simple and plausible, though not rigorous, way from the Gorter-Casimir two-fluid model.^{5,6} On that model, the density of superconducting electrons at reduced temperature $t = T/T_c$ (where T_c = the transition temperature) is $n_s/n_0 = 1 - t^4$. This

 ¹ Bardeen, Cooper, and Schrieffer, Phys. Rev. 108, 1175 (1957).
 ² C. S. Whitehead, Proc. Roy. Soc. (London) A238, 175 (1956).
 ³ R. E. Glover, III, and M. Tinkham, Phys. Rev. 108, 243 (1957); M. Tinkham, Phys. Rev. 110, 26 (1958).
 ⁴ A. L. Schawlow, Phys. Rev. 109, 1856 (1958).

⁵ Daunt, Miller, Pippard, and Shoenberg, Phys. Rev. 74, 843

^{(1948).} ⁶ D. Shoenberg, *Superconductivity* (Cambridge University Press, Cambridge, 1952), Chaps. 5 and 6.

follows thermodynamically if the threshold field is accurately parabolic, i.e.,

$$H_c = H_0(1-t^2),$$

where H_c is the critical magnetic field at temperature t. If these superconducting electrons have an effective mass m^* , and have been accelerated by the field without collisions, the penetration depth is

and so

$$\lambda = \lambda_0 / (1 - t^4)^{\frac{1}{2}} = \lambda_0 \gamma,$$

 $\lambda = (m^*/4n_s e^2)^{\frac{1}{2}} \propto 1/(n_s)^{\frac{1}{2}},$

where λ_0 = penetration depth at absolute zero and

$$y = 1/(1-t^4)^{\frac{1}{2}}$$

The experimentally observed temperature variation of the penetration depth fits this curve quite well.⁵⁻⁷

But there is now considerable evidence that the critical field curve is really not quite parabolic^{8,9} because of the existence of a gap in the electron energy spectrum. Lewis¹⁰ extended the two-fluid model to include an energy gap independent of temperature. Figure 1 shows the values of λ on this model relative to those without the gap. It is seen that λ falls more rapidly as one goes down in temperature because of the gap. In Fig. 2 the values of λ are replotted agianst $y=1/(1-t^4)^{\frac{1}{2}}$. Now it is seen that near the transition temperature, T_c , the very rapid variation of y with t dominates the temperature dependence. Thus in Fig. 2 the graph is very nearly a straight line for most of its length, whether there is an energy gap or not. Only at low temperatures where y is not varying so rapidly



FIG. 1. Temperature dependence of the superconducting penetration depth. (a) Gorter-Casimir two-fluid model; (b) Lewis' two-fluid model with energy gap.

⁷ R. G. Chambers, Proc. Cambridge Phil. Soc. **52**, 363 (1956). ⁸ B. Serin, in *Progress in Low-Temperature Physics*, edited by C. Gorter (Interscience Publishers, Inc., New York, 1955),

Vol. 1, Chap. 7. ⁹ W. S. Corak and C. B. Satterthwaite, Phys. Rev. **102**, 662

(1956). 10 H W Lowin Dhys Day 102 1508 (1056)





FIG. 2. Superconducting penetration depth as a function of $y=1/(1-t^{4})^{\frac{1}{2}}$ with energy gap (Lewis) and without gap (Gorter-Casimir).

with T, does the slope of the penetration depth curve change appreciably because of the gap. Since we do not know λ_0 in advance, we can only distinguish these if we can measure the change of λ with temperature accurately enough to show this curvature.

The fundamental superconductivity theory of Bardeen, Cooper, and Schrieffer¹ also derives and uses an energy gap, although one which vanishes at the transition temperature. Over the region in which the relation between λ and T shows appreciable curvature, the gap is substantially constant. Thus the BCS theory predicts much the same sort of a departure from the straight line when λ is plotted against y. The differences arising from the very different details of the two theories are in this case quantitative, not qualitative.

Any theory with a finite energy gap must give this sort of behavior. If the number of normal electrons varies according to some power of t, such as t^4 , at high temperatures, it must fall more rapidly than the power law at lower temperatures. This follows because the probability of excitation across a gap is an exponential function of 1/t, which can be approximated by a power law only for small values of the argument.

The effects of the energy gap on penetration depth have, up to now, not been observed because earlier experiments did not have enough sensitivity to permit them to study the low-temperature region. The total change expected from 0.9 T_c to absolute zero is only $0.6 \lambda_0$, or about 300 angstroms. The apparatus described here has sufficient sensitivity to see a change of about 4 angstroms, so that these effects are quite readily detectable and measurable.

II. METHOD

The superconducting sample is a long single-crystal rod (length 17 cm, diameter 0.74 cm) which serves as

so that



FIG. 3. Oscillator circuit for penetration depth measurement.

the core of a solenoid. The inductance of the solenoid is proportional to the cross-sectional area of the space occupied by flux, that is, the space between the coil and the sample plus whatever distance the flux penetrates into the surface of the sample. As the temperature is changed, the penetration depth changes, and the inductance changes with it. Then

change in inductance

total inductance

change in penetration depth×rod circumference

total cross section occupied by flux

To make the inductance change measurable with sufficient sensitivity, the coil is resonated by a mica capacitor, C_1 to some frequency around 100 kc/sec. The combination forms the tank circuit of an oscillator, as shown in Fig. 3. When the inductance, L, alters, because of a change in penetration depth, the oscillator frequency shifts:

so that

$$2\pi r\delta\lambda/A = \delta L/L = 2\delta f/f$$

 $\delta f/f = \frac{1}{2} \delta L/L$,

or

$$\delta \lambda = (A/\pi r) \delta f/f,$$

where r is the radius of the sample rod and A is the cross-sectional area between the rod and the coil.

If A is not large it may be hard to measure accurately, but it can be eliminated by calibrating with several samples of different diameters. Since L is proportional to $A = \pi r_0^2 - \pi r^2$, where r_0 and r are the effective radii of the coil and core, respectively, then

$$A \alpha L \alpha 1/f^2 = B + Cr^2.$$

Thus a graph of $1/f^2$ against r^2 should be, and is, a straight line. Then

$$d\lambda = dr = -(1/Br)(1/f^2)(df/f),$$

 $-(1/f^3)df = Brdr,$

and the constant B is obtained from the intercept of the $1/f^2$ against r^2 plot, or indeed simply by measuring the resonant frequency with an empty coil. For highest sensitivity, A should be small, that is the coil should be close to the sample. It cannot, however, be wound directly on the sample, as that would surely strain the surface severely and radically alter the penetration depth. In preliminary work, the coil consisted of two layers of No. 44 copper wire would on a glass tube inside which the sample was placed. With this arrangement, the radial distance between the coil and sample could hardly be made less than about a millimeter.

About five times closer spacing was obtained by using a coil wound inside the glass tube, rather than outside.¹¹ The coil was wound on a steel mandrel, with paper shims separating it from the mandrel. It was coated with an epoxy resin to give it rigidity, baked, and removed from the mandrel. After a second coating of epoxy resin, it was slipped inside the glass tube and baked again. The resultant coil was stable and rigid, although differences in thermal expansion would eventually break it loose after repeated coolings.

With the small spacing made possible by the insidewound coil, the inductance was so low that copper coils had a Q too low to permit oscillation, and so a superconducting coil of 0.003-in. diameter Formvarcoated niobium wire was used. Then very stable oscillations were obtained right up to the transition temperature of the sample, but not above it. The outsidewound copper coil was used to measure the transition temperature, because its higher inductance permitted oscillations to be sustained even when the rod was in the normal state.

The superconducting coil was adopted with some reluctance, because it seemed possible in principle that changes of the niobium penetration depth with temperature might occur and give misleading results. If the penetration depth varied as $1/(1-t^4)^{\frac{1}{2}}$, the changes would be indeed negligible in the region of our experiment which is far below the transition temperature of niobium (8°K). However, as a check, tests were run with a dummy sample consisting of a polycrystalline rod of lead. Over the range from 3.5°K to 1.4°K the oscillator frequency did not fluctuate by more than ± 0.3 cps and at 1.4°K it was within 0.2 cps of the frequency at 3.5°K. A shift of the order of 0.4 cps might have been expected from the small variation of λ for lead over our temperature range. As the observed variation is small, we may safely neglect in this region

 $^{^{11}}$ We are very much indebted to Mr. A. G. Olson of the Bell Telephone Laboratories Coil Shop for devising the procedure and for winding these coils.

any effects due to temperature-dependent penetration into the coil. We can thus take advantage of the greater sensitivity and stability made possible by the superconducting coil. At higher temperatures it would be necessary to use a copper coil or to allow for temperature-dependent penetration into the niobium coil.

In the oscillator circuit of Fig. 3, the amplitude can be adjusted by changing the feedback resistor R_1 , and by adjusting the plate voltage. With the niobium coil, a plate voltage of 35 volts and a feedback resistance of 1.5 megohms gives an amplitude of about 0.1 volt. This corresponds to an alternating field of about 0.6 oe rms. All data used were taken at least 0.03°K from the transition temperature, i.e., where the sample had a critical field of at least 6 oersteds and usually very much more. For measuring the transition temperature with the copper coil, still smaller field amplitudes were used.

The earth's magnetic field was neutralized by two pairs of Helmholtz coils to within about two percent. The importance of this neutralization has been pointed out by Laurmann and Shoenberg¹² and by Chambers.⁷ However, the stray field is found to have much less effect in these experiments than in their work. This is probably due to the nearly complete Meissner effect and the extremely high conductivity of our very pure tin specimens.¹³ At this frequency, inclusions of normal flux would be immobilized by eddy-current damping.

The operating frequency is chosen to be low enough so that the skin depth is very much greater than the superconducting penetration depth. In fact, the experiment to determine the transition temperature gives us at the same time the total inductance change between the normal and superconducting states, and so the normal-state skin depth. The skin depth determined in this way is 6.90×10^{-4} cm or about 140 times the superconducting penetration depth at absolute zero. Thus, in the superconducting state, the penetration depth is determined solely by the superelectrons, and no allowance need be made for high-frequency screening by normal electrons. Moreover, the frequency used is low enough so that $h\nu$ is very much less than kT, and effects due to the proximity of induced transitions across the gap, which might be troublesome with microwaves, are indeed negligible.

If the skin depth is proportional to (frequency)⁻¹, as it should be for the anomalous skin effect, at 1000 Mc/sec it would be 3.3×10^{-5} cm, while at 100 000 Mc/sec, it would be 7.0×10^{-6} cm. At neither frequency is it large enough so that the effects of normal electrons can be ignored, particularly near the transition temperature, where λ is large. Nevertheless, it is sometimes assumed, even at the highest frequency, that the field distribution is determined solely by superelectrons. It may be safe to make this assumption for sufficiently low temperatures, but present theories are not a safe guide to how low is sufficient.

The oscillator frequency is high enough, on the other hand, to be easily measurable with great precision in a short time. Frequency measurements were made with a Berkeley events-per-unit-time counter, which counted cycles for exactly ten seconds. In this way, frequency was measured to 0.1 cps, and within a minute four or five such readings could be taken. Constancy of these successive readings within a count or so was good evidence for the attainment of thermal equilibrium in the sample. If the temperature were not changed, the frequency of the oscillator would usually hold constant within one or two counts (parts per million) over an hour or more.

The samples were single-crystal rods 17 cm long, 0.74 cm in diameter, of tin grown in graphite molds open at the ends to a dental plaster boat containing a seed of the desired orientation. After the crystal was grown, the mold was removed and the sample was etched in dilute hydrochloric acid to check its orientation. Finally the sample was chemically polished¹⁴ in freshly mixed aqua regia at 100°C, and then hung vertically until used. This final precaution was quite necessary, as these pure crystals are very soft, and if tilted from the vertical without support will slip easily under their own weight. Only the center two inches of sample is surrounded by the coil, the remainder serving to eliminate end effects from the measurements. Such handling as could not be avoided was done at the ends. During measurements these ends are fastened to the glass tube with cheese wax to prevent movement, although space is left for helium to circulate between the coil and the sample.

The capacitance between the niobium coil and the sample might be as large as 50 micromicrofarads, which is less than $\frac{1}{10}\%$ of the tuning capacitance of 72 000 micromicrofarads. The effective spacing between coil and sample might conceivably vary with temperature as much as the magnetic penetration varies. The resulting capacitance change would shift the oscillator frequency only $\frac{1}{10}\%$ as much as would the accompanying inductance change. Even the changes of the dielectric constant of helium with temperature over the whole range of the experiment would change the oscillator frequency by only one part per million, which is just about our limit of resolution. Thus the stray capacitance can be safely neglected.

This method is related to that proposed originally by Casimir¹⁵ and realized by Laurmann and Shoenberg.¹² It, and the method of Chambers,⁷ differs from theirs in that we measure changes in a self-inductance rather than a mutual inductance. The oscillator frequency shift has not previously been applied to penetration

¹² E. Laurmann and D. Shoenberg, Proc. Roy. Soc. (London) A198, 560 (1949).

¹³ Spec-pure tin from Vulcan Detinning Company, Sewaren, New Jersey.

¹⁴ The smoothness of the chemically polished surfaces was confirmed by electron micrographs made by C. J. Calbick.
¹⁵ H. B. G. Casimir, Physica 7, 887 (1940).



FIG. 4. Transition of tin crystal from normal to superconducting state as shown by oscillator frequency shift.

depth measurements, but it is related to the ancient beat frequency method of measuring capacitance and inductance but with the counter replacing the heterodyne comparison oscillator. In a sense, these resonant methods balance the inductive reactance by a capacitive reactance at the resonant frequency. Unlike the bridge method, substantially all the balance circuit is in the liquid helium. This is one of the factors permitting the attainment of high sensitivity.

This method is also very suitable for studying the reactive skin depth, or the surface reactance, in the normal state. In particular, we have applied it in a preliminary way to some studies of magnetoresistance. We have noted a peculiar decrease in skin depth with magnetic fields up to about a thousand oersteds which is associated with the anomalous nature of conduction in high-purity samples even at these frequencies.



FIG. 5. Temperature dependence of oscillator frequency and penetration depth for tin crystal with transverse c axis (sample Sn121). Theoretical curve from theory of Bardeen, Cooper, and Schrieffer.

Transition Curves

Figure 4 shows the transition curve, in a copper coil, of a tin crystal rod. From this sharp transition, in which the graph by no means does justice to the fineness of the frequency shift data, we can determine that $T_c = 3.7205$ °K on the T_{55E} scale¹⁶ (3.722°K on the 1948 scale). T_c here is taken at the midpoint of the steep transition curve.¹² Both specimens with a longitudinal fourfold crystal axis and with a transverse fourfold axis had the same transition temperature within our accuracy which is about 0.0005°K.

The tail at lower temperatures in Fig. 4 is not due to breadth in the transition, but to the change in λ with temperature. From this portion of the data we find for sample Sn121 (c axis $_{1}$ rod axis) df/dy = 2.6(5) cps/unit of y, where $y=1/[1-(T/T_c)^4]^{\frac{3}{2}}$, and since B for this particular coil is -1.17×10^{-9} , then $d\lambda/dy=5.2 \times 10^{-6}$ cm. This is the quantity which would on the two-fluid



FIG. 6. Temperature dependence of oscillator frequency and penetration depth for tin crystal with transverse c axis (sample Sn121) for low temperatures.

model have been identified as λ_0 , and is in good agreement with several earlier determinations by various methods.12,17,18

A sample with the crystalline fourfold axis (c axis) along the rod axis gave very similar results, but an accurate determination of $\delta\lambda/\delta y$ was deferred until the experiments with the niobium coil.

Also from the transition curves we can get the total shift from the normal to the superconducting state. and so derive the skin depth in the normal state. In this way, for Sn120 (c axis $_{II}$ rod axis), the skin depth, δ at 102 kc/sec is 7.18×10⁻⁴ cm; and for Sn121 (axis $_{\perp}$ rod axis), $\delta = 6.90 \times 10^{-4}$ cm. Both of these, as well as the values of $d\lambda/dy$ above, are averages for current flow around the rod.

¹⁸ J. M. Lock, Proc. Roy. Soc. (London) A208, 391 (1951).

¹⁶ J. R. Clement, "Liquid helium temperature scale," Naval

 ¹⁷ A. B. Pippard, Proc. Roy. Soc. (London) A191, 399 (1947), recalculated in T. E. Faber and A. B. Pippard, Proc. Roy. Soc. (London) A231, 336 (1955).

TEMPERATURE DEPENDENCE OF THE PENETRATION DEPTH

In Fig. 5, is shown the variation in oscillator frequency, and from it the penetration depth with temperature for Sn121 measured in a niobium coil. For this coil and sample $B=5.74\times10^{-9}$, and so 0.1-cps shift in oscillator frequency corresponds to 4.04 angstroms change in penetration depth. Since we do not know λ_0 in advance, and only measure $d\lambda/dt$, the λ scale has an additive constant which can be adjusted arbitrarily, although we are not free to stretch or contract the scale.

From Fig. 5, it is seen that λ varies quite linearly with y from about y=1.8 to y=6, i.e., from 3.4° K to 3.696° K. At lower temperatures there is a definite curvature in the direction predicted by the energy gap theories. To show this more clearly, Fig. 6 shows the portion of the curve from y=1 to y=2.

Since we do not directly measure λ in these experiments, but only its changes with temperature, we can



FIG. 7. $d\lambda/dy$ as a function of y for sample Sn121. Theoretical curve from Bardeen, Cooper, and Schrieffer.

move the experimental points up or down the λ axis by adding to all of them any fixed quantity. We have arbitrarily chosen to match them to the BCS theoretical curve at 0°K (y=1); a better over-all fit could have been obtained by matching at some intermediate point. The over-all agreement is excellent and the experiments do show a deviation from the straight line plot very much like that predicted as a consequence of the energy gap.

An even more stringent comparison between experiment and theory is given in Fig. 7, where $d\lambda/dy$ is plotted against y. Again the agreement is quite good, and this is especially remarkable in view of the fact that there are *no adjustable parameters*. On the Gorter-Casimir theory, the graph would be a horizontal straight line.

Figures 8 and 9 show, respectively, λ and $d\lambda/dy$ for another specimen (Sn120) with the *c* axis along the rod axis. These data are probably a little less accurate than those for Sn121, because refinements were made



FIG. 8. Temperature dependence of penetration depth for tin crystal with c axis parallel to rod axis (sample Sn120).

in the temperature measuring procedure after they were taken. However, the agreement with theory is still very good.

Such small discrepancies as remain may be due to uncertainty in the input data for the theoretical calculation (see Table IV of reference 1) or to approximations in it. At any rate, the agreement is very satisfactory.

It is interesting to note that the slope of the experimental and theoretical curves for y > 1.7 or so is about 520 A per unit of y, in excellent agreement with our copper coil results and with those of previous experimenters. If one were to assume, as earlier investigators have done, that λ is proportional to y, then one would deduce $\lambda_0 = d\lambda/dy = 520$ angstroms. However, because of the hook in the curve at small y, this assumption is no longer valid, and we cannot really deduce λ_0 from the experiments at all. The best we can do is assume the approximate validity of the BCS theory (or some other). The BCS theory gives $\lambda_0 = 567$ A, and since this theory fits the data quite well, that value is probably reasonably good. However, the absolute values of λ are all greater than $y\lambda_0$, by as much as 200 A for the theoretical values, or 100 A for our experiment. This might be important in studies of thin films of colloids, where λ becomes comparable with the sample dimensions.^{2–4} In such experiments the effective value of λ was found to be greater than the value which was thought to be characteristic of a bulk super-



FIG. 9. $d\lambda/dy$ as a function of y for sample Sn120.



FIG. 10. Temperature dependence of penetration depth for sample Sn121 compared with Lewis' extended two-fluid model with energy gap.

conductor, presumably because of the small dimension of the sample. If, however, the values of λ for bulk superconductors are larger than assumed previously, part, but not all, of the change attributed to finite particle size is removed.

We may also compare our experimental results with the phenomenological energy gap theory of Lewis. There λ_0 is an adjustable parameter, and we choose it to fit the slope from y=2 to y=5. In this way, we obtain $\lambda_0=425$ A. Again, in Fig. 10 we arbitrarily fit the experimental points to the theoretical at 0°K (y=1). The theoretical curve has an increased slope at small y, but not as much increase as indicated by our experiments or the BCS theory. The slope is plotted in Fig. 11. With the extra adjustable parameter (λ_0), the curve fits our data almost as well as those of the BCS theory, although the slope at small y is definitely less. Actually, one would not really expect the fixed energy gap alone to fit experiments well except at extremely low temperature, lower than those of these experiments.

CONCLUSIONS

The dependence of penetration depth on temperature, when observed with sufficient sensitivity, shows the effect of an energy gap in the spectrum of electron excitations. The experimental results agree well with the theory of Bardeen, Cooper, and Schrieffer, but can also be fitted fairly well to the extended two-fluid phenomenological theory of Lewis.

The method would be equally applicable to a metal

such as mercury, where the effects of an energy gap on specific heat or critical field have not yet been observed. It is suspected that in mercury the lattice specific heat varies in a complicated way with temperature, because of the low Debye temperature. This lattice specific heat must be subtracted before the electronic term can be isolated, and so the effect of a gap is masked. No such masking should disturb the penetration measurements which are directly affected by the electron excitation.

We do not observe any appreciable anisotropy of the penetration depth. Indeed, if the anisotropy follows that found with microwaves by Pippard¹⁹ and Fawcett,²⁰ none should be observed with specimens in which the current flows around the diameter of a rod. In so doing,



FIG. 11. $d\lambda/dy$ for Sn121 compared with Lewis' theory.

the current must traverse several crystal directions, and so our value is always an average.

However, our method can be modified to use flat specimens and then we should be able to study any anisotropy without the grave complications of microwave methods. This work is being undertaken.

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 ¹⁹ A. B. Pippard, Proc. Roy. Soc. (London) A203, 98 (1950).
 ²⁰ E. Fawcett, Proc. Roy. Soc. (London) A232, 519 (1955).