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An Experimental Examination of the Electrostatic Behaviour of Supraconductors

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1—INTRODUCTION

In previous papers of F. and H. London* supraconductivity has been described as a phenomenon, in which the current density is not connected with the electric field, as in normal conductors, but depends on the magnetic field strength according to the equation

$$\Lambda c \operatorname{curl} \mathrm{J} = -\mathrm{H} \tag{1}$$

with

$$\mathrm{B} = \mathrm{H}$$

and with

$$\Lambda = m/ne^2,$$

a new characteristic constant which contains the number n of supraconducting electrons.

The behaviour of the *electric* field is not completely determined by this equation. Using Maxwell's induction law one can conclude from (1) only that

$$\Lambda c \operatorname{curl} \dot{\mathrm{J}} = c \operatorname{curl} \mathrm{E}$$

or

$$\Lambda \dot{\mathrm{J}} = \mathrm{E} + \operatorname{grad} \mu, \tag{2}$$

where the physical significance of grad μ is yet unknown.

Two alternative hypotheses have been made, namely

$$\operatorname{grad} \mu = 0 \quad \text{and} \quad \mu = -\Lambda c^2 \rho.$$

The first assumption excludes any stationary fields by the equation

$$\mathrm{E} = \Lambda \dot{\mathrm{J}} \tag{3}$$

and gives surface charges wherever the electric field of the external medium has a normal component.

* 'Proc. Roy. Soc.,' A, vol. 149, p. 71 (1935), referred to as A; 'Physica,' vol. 2, p. 341 (1935); F. London, 'Proc. Roy. Soc.,' A, vol. 152, p. 24 (1935).

The second assumption is distinguished so far as the resulting equation

$$\Lambda\,(\dot{\mathrm{J}} + c^2 \operatorname{grad} \rho) = \mathrm{E} \tag{4}$$

can be combined with (1) in a single law by the four dimensional representation

$$\Lambda\left(\frac{\partial \mathrm{J}_i}{\partial x_k} - \frac{\partial \mathrm{J}_k}{\partial x_i}\right) = \frac{1}{c} f_{ik},$$

with $\mathrm{J}_1, \mathrm{J}_2, \mathrm{J}_3, \mathrm{J}_4 = \mathrm{J}_x, \mathrm{J}_y, \mathrm{J}_z, ic\rho$; $x_1, x_2, x_3, x_4 = x, y, x, ict$; and $if_{14}, if_{24}, if_{34}$; $f_{23}, f_{31}, f_{12} = \mathrm{E}_x, \mathrm{E}_y, \mathrm{E}_z$; $\mathrm{H}_x, \mathrm{H}_y, \mathrm{H}_z$.

In this description the electric and the magnetic behaviour are quite symmetrical. Both fields penetrate the supraconductor in a thin layer where they vanish exponentially, so that in a distance

$$d = \sqrt{\Lambda c^2} = \sqrt{\frac{mc^2}{ne^2}}, \tag{5}$$

they are reduced to the e-th part of their surface values; d is probably of the order of 10^{-6} to 10^{-4} cm. In this way the macroscopic result $\mathrm{E} = 0$, $\mathrm{B} = 0$, or $f_{ik} = 0$, can be represented. Equation (4), which was completed by the simplifying assumption $\mathrm{E} = \mathrm{D}$, still requires a boundary condition. With respect to the boundary, supraconductor–normal conductor, it has been shown by an energy consideration that $\rho = 0$ must be postulated.* This leads generally to a discontinuity of the normal component of E, as with two adjacent normal conductors. At the boundary, supraconductor–insulator, however, a continuous D_n, *i.e.*, exclusion of surface charges, seemed to be a plausible boundary condition, analogous to that of two adjacent insulators. For, as a consequence of (4), one obtains an energy density of the value $\frac{1}{2}\Lambda c^2 \rho^2$ attributed to the space charges ρ† and this would become infinite in a surface charge. If, nevertheless, surface charges could be formed without any peculiar energy, the arguments given for the boundary with a normal conductor would apply at this boundary too. Accordingly $\rho = 0$ would hold at the whole surface of the supraconductor and therefore everywhere within it, and no substantial difference between equations (3) and (4) would remain.

Both equation (4) with continuous D_n and equation (3) are in agreement with all experiments carried out hitherto with pure supraconducting phases. A method of deciding between them is the measurement of the

* M. v. Laue, F. and H. London, ' Z. Physik,' vol. 96, p. 359 (1935).

† A, p. 77.

capacity of a condenser with supraconducting plates. If D_n should be continuous, the capacity would decrease when the condenser was cooled below the transition temperature, and an elementary calculation shows that the change would be equivalent to an increase of the distance between the two plates by $2d$ (*in vacuo*).

Such a measurement has to be carried out with a sufficiently low measuring voltage, as there could exist a threshold value D_{nT} for the normal component of the electric induction above which the surface of the supraconductor would become normal conducting. Then the lines of electric induction could terminate discontinuously in surface charges, which are excluded only for supraconductors. The electric field inside the supraconductor would disappear and accordingly the field energy would decrease. It is of course impossible to give a definite figure for this electric threshold value as one does not know what energy the formation of a " normal conducting surface " would require. It would be fairly high, if a real normal conducting phase of at least the atomic extension a had to be formed, namely $D_{nT} \approx \sqrt{a/d} \,.\, H_T$, where H_T is the magnetic threshold value. For $H_T = 330$ gauss and $a/d = 10^{-4}$ one would obtain 3·3 electrostatic units or 10^3 volts cm^{-1}. As a precaution, however, an estimation has also been made on the assumption that not even a real phase transformation might be necessary, but that it might be sufficient that as many electrons become normal conducting as are required to terminate the lines of induction of the external field. From this a lowest limit for the threshold value of the order

$$D_{nT} = \frac{1}{\sqrt{nmc^2}} \,.\, H_T^2$$

follows above which the change in capacity would become inversely proportional to the measuring voltage. The greatest number n of supraconducting electrons which can reasonably be assumed is the number of valency electrons. For mercury, with 2 valency electrons per atom at a temperature of 1·9° K, where $H_T = 330$ gauss,* one gets

$$D_{nT} = 1 \cdot 7 \times 10^{-2} \text{ volts cm}^{-1}.$$

On the other hand the effect would disappear if the surface of the supraconductor were covered with normal conducting impurities. Also disturbance of the surface by polishing or oxidation might impair the effect. Therefore a natural surface of high purity was required.

* ' Comm. Phys. Lab. Leiden,' No. 180*d* (1926).

2—Method

The Condenser

Mercury seemed to be the most suitable substance, as it can easily be purified, so that it shows a 100% Meissner-effect.* The condenser was made by distilling in the mercury in a high vacuum. The container for the mercury had a shape shown in fig. 1. Two chambers were formed by two half-cylinders of glass tube closed at the bottom between which a mica foil M, 18 μ thick, was clamped. The parts were kept in position by wires wound around the glass half-cylinders. The container was placed in a wider glass vessel which was connected with the distillation apparatus. As current leads two platinum wires Pt were sealed through the outer vessel. The first drops of mercury, which are usually of lower purity were distilled into another vessel before the condenser was connected. The mercury used was "distilled mercury" of Messrs. Hopkin & Williams which had been purified by electrolysis before it was distilled into the condenser. The distillation temperature was 100° C. After the distillation the vessel was filled with pure helium, to give heat contact, and then it was sealed off at S. Reaction of the platinum leads with the mercury was minimized by freezing the mercury soon after the distillation finished, so that the platinum was in contact with liquid mercury for not longer than one day.

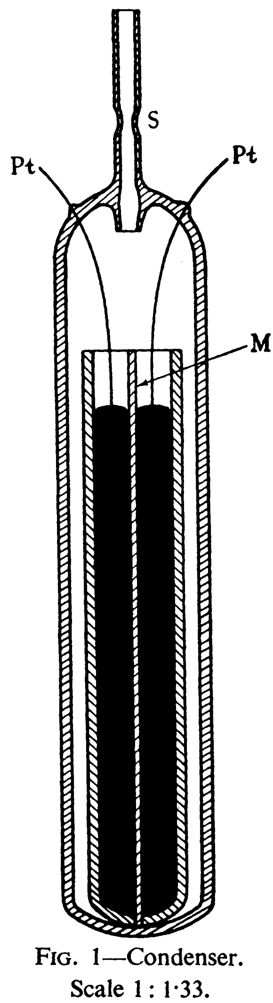

Fig. 1—Condenser. Scale 1 : 1·33.

For a proper freezing two precautions were necessary. The condenser had to be cooled slowly from the bottom in order to avoid the formation of hollow spaces in the solid mercury. Secondly the mercury had to be prevented from sticking at the glass walls because otherwise it would sever from the mica or even split it owing to its high thermal contraction. To avoid this, acid cleaned filter paper was placed at the inner surface of the glass half-cylinders.

* Keeley, Mendelssohn, and Moore, 'Nature,' vol. 134, p. 773 (1934).

Still a small separation of the mercury took place, which could be seen from a fall in capacity larger than would follow from thermal contraction alone. This was considered in the evaluation of the relative capacity change by assuming the most unfavourable case that this separation was spread continuously over the surface, while in the calculation of the actual measuring field-strength the original plate distance was assumed. The reduction in the relative sensitivity caused by this separation was 25%; the plate distance was equivalent to $3 \cdot 8\ \mu$ for the plates of a condenser with vacuum dielectric and the same capacity per cm^2.

The Electric Measurement

The problem was to measure a comparatively large change in capacity ($0 \cdot 1$%) by a very low measuring voltage, so that the change in voltage produced by the change in capacity would be of the order of 10^{-8} volts.

For the measurement alternating current of 150,000 cycles per second was produced by a small valve generator and transformed down by two ironless transformers, the secondary coil of the first one being connected with the primary coil of the second. The ratio of the two transformers was known and their reactance was high compared with the subsequent load-impedance. Thus, by measuring the voltage on the input side the voltage applied to the mercury condenser could be determined. It was about 10^{-5} volt.

At the frequency used the change in reactance and resistance occurring at the transition into the supraconducting state are still negligible compared with the change in capacitance to be detected so that such a measurement can be considered to be quasistatic.

The capacity was measured by a bridge of which all four branches consisted of condensers, as the bridge was part of a tuned circuit of the amplifier which would have been damped too much if a resistance-capacity bridge had been used. In fig. 2 C_x is the mercury condenser, C_v a calibrated variable condenser, by which a change in C_x could be balanced. The compensation of the dielectric losses was carried out by a small variable mutual inductance M with resistances R_1, R_2 in its primary circuit, which were large compared with the reactance of the primary coil. By this method the inconvenience of using a variable resistance becomes unnecessary. The condensers C_1, C_2 and the high ohmic resistance R_3, R_4 were used to protect the condenser from potential differences due to thermo-forces. For this purpose R_4 was cooled together with C_x; R_3 was also cooled in order to compensate for all variations of R_4.

The amplifier worked on the heterodyne principle: After a screened-grid high frequency stage an auxiliary oscillation was superimposed, which was just strong enough to give sufficient input to the detector valve, which followed. With this precaution and by the screening effect of the first valve, a reaction of the auxiliary voltage back to the mercury-

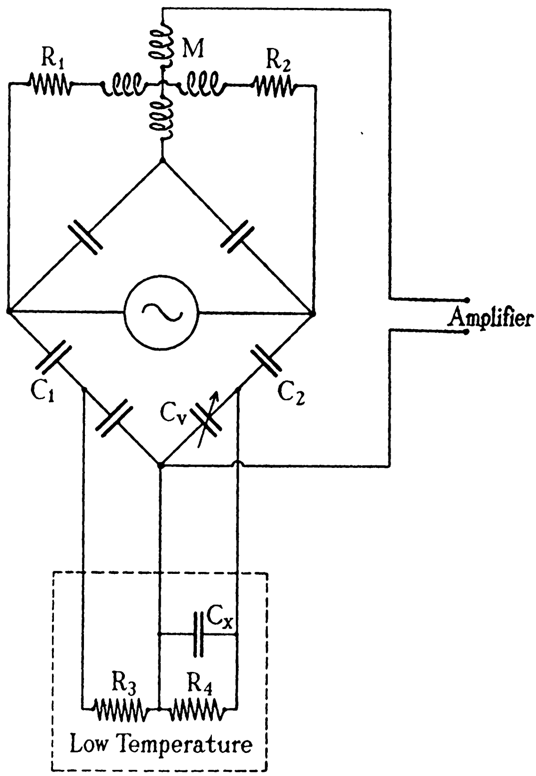

FIG. 2—The bridge circuit.

condenser could be avoided, which might otherwise have increased the measuring voltage. The audible beat frequency was amplified by a 4-valve low-frequency amplifier. One stage of it had a tuned iron-cored transformer, which was provided with reaction. By means of a variable load resistance this reaction could be adjusted, so that the circuit was almost on the point of oscillation, and thus a selectivity of about 2 cycles per second could be obtained. This was necessary in order to cut down the background noise. In order to get the beat-frequency sufficiently constant, the two high-frequency oscillators were designed and operated in exactly the same way.

The Low Temperature Arrangement

The low temperature was produced by a small helium liquefier, working by the expansion method,* and measured by a vapour pressure thermometer. The condenser, together with the resistances R_3, R_4, was in a vessel soldered at the bottom of the helium container; for thermal contact the vessel was filled with helium gas. That the mercury had attained the temperature of the liquefier was ascertained by measuring its magnetic threshold value.

For this purpose the condenser formed the core of a mutual inductance the secondary coil of which was connected to a ballistic galvanometer. In this way the change in the magnetic flux through the mercury could be observed, when a magnetic field increasing by steps was produced by the primary coil. As long as the mercury was supraconductive, this flux remained zero; when the threshold field was exceeded, it changed to its normal value producing a deflexion of the galvanometer. On decreasing the field again below the threshold value the opposite change in flux took place, according to the Meissner-effect.†

As even in the temperature region of liquid helium the capacity of the condenser changed a little with temperature owing to the mentioned separation of the mercury from the mica due to thermal contraction, only comparative measurements at constant temperature were carried out, the normal conductivity being restored by a magnetic field.

3—Results

At a temperature of $1\cdot 8_5^\circ$ K no change in capacity of the mercury condenser was observed when the supraconductivity was destroyed by a magnetic field. Expressing all capacities in terms of the plate distance δ, which a vacuum condenser of equal capacity and area would have, the limit of error corresponded to a change in δ of $4\cdot 6 \times 10^{-7}$ cm, or of $2\cdot 3 \times 10^{-7}$ cm for either plate. Since the least favourable value of d, the penetration depth, calculated from the assumption of 2 supraconducting electrons per atom by equation (5), is $1\cdot 8 \times 10^{-6}$ cm, the limit of error is less than 13% of the effect. The peak-value of the electric induction of the measuring field was less than $3\cdot 9 \times 10^{-2}$ volts cm^{-1}, while the lowest limit of the threshold induction was $D_{nT} = 1\cdot 7 \times 10^{-2}$ volts cm^{-1} (*see*

* Simon, 'Phys. Z.,' vol. 34, p. 232 (1933); Simon and Ahlberg, 'Z. Physik,' vol. 81, p. 816 (1933).

† A similar arrangement was first used by Rjabinin and Shubnikow, 'Phys. Z. Sowjet,' vol. 5, p. 641 (1934).

p. 3). It has therefore been exceeded during a part of the period. But even then a change in δ of 2×10^{-6} cm ought to occur, which is still more than four times the limit of error. Measurements have also been carried out with a higher measuring voltage and accordingly with a higher exactness of the capacity measurement. At an induction of 10 volts cm^{-1} the change in the equivalent vacuum distance δ was found to be less than $7 \cdot 2 \times 10^{-8}$ cm, *i.e.*, $3 \cdot 6 \times 10^{-8}$ cm for a single plate, or less than 2% of the effect.

4—Conclusions

It follows from these measurements that no electrostatic fields exist in a pure supraconductor, not even in a thin surface layer, at least to the approximation to which this is true for normal conductors. Accordingly we shall assume grad $\mu = 0$ in equation (2) and consider

$$\Lambda c \text{ curl J} = -\text{H} \tag{1}$$

$$\Lambda \dot{\text{J}} = \text{E} \tag{3}$$

as the most simple description of the behaviour of a supraconductor. In all stationary cases the surface of a supraconductor is therefore exactly an equipotential surface. This gives the boundary condition for the electric field.*

Equations (1) and (3) can be considered as valid for a single supraconducting phase and also for phase transitions in a magnetic field.† However, it is not possible to treat the transition curve of a supraconducting wire with a current maintained from outside as an equilibrium

* Consequently a supraconducting sphere in a homogeneous electrostatic field behaves exactly like a normal conducting one, without the small deviations given in A, p. 79, for a continuous D_n. The absolute value of the *current* through a supraconducting sphere, which is determined by the electric field in the adjacent normal conductor, is also slightly different from that previously given (A, p. 80). The constant k is now given by

$$k = \frac{3\sigma \text{ER}}{\sinh \beta\text{R}} \cdot \frac{\beta^2\text{R}^2}{1 - \beta\text{R} \operatorname{ctgh} \beta\text{R}}$$

instead of

$$k = \frac{3\sigma \text{ER}}{\sinh \beta\text{R}}\left[2 + \frac{\beta^2\text{R}^2}{1 - \beta\text{R} \operatorname{ctgh} \beta\text{R}}\right].$$

The *distribution* of the current remains unchanged. (The result for the current followed already from the theoretical consideration of von Laue, F. and H. London, 'Z. Phys.,' *loc. cit.*)

† H. London, 'Proc. Roy. Soc.,' A, vol. 152, p. 650 (1935).

problem of two separated phases.* It is probable that in this case a homogeneous supraconducting phase does not exist at all.

I should like to express my thanks to Professor F. A. Lindemann, F.R.S., for his generous hospitality in the Clarendon Laboratory.

My thanks are due to Professor F. Simon for putting one of his helium liquefiers at my disposal and for his helpful interest in my work.

Finally I am indebted to Mr. A. H. Cooke, Dr. N. Kürti, and Mr. G. L. Pickard for their kind help during the experiments.

Summary

The question, whether, in a supraconductor, the lines of electric induction terminate discontinuously in surface charges or whether they penetrate a thin layer of the supraconductor was undecided. It has been decided experimentally in favour of the surface charges by the measurement of the capacity of a supraconducting condenser. Accordingly $E = 0$ is valid in stationary conditions even in surface regions of the order of magnitude of 10^{-7} cm.

The measurements were carried out with a very low measuring voltage in order to exclude disturbance of supraconductivity due to a possible electric threshold value.

* Equation (4) gave such a possibility, though not without introducing new hypothetical assumptions (A, p. 80, ff.).