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TUNNEL EFFECT. -

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1. - INTRODUCTION. -

In the present work we have made a new analysis of the physical properties of a type I superconducting cylinder. The calculation are developed in the following manner. In the first paragraph is calculated the dependence of the order parameter on the external magnetic field and on the quantum number n . The expression for the induced current density, as a function of the order parameter and thus as a function of the external magnetic field and of the quantum n , is found in the second paragraph. Finally, in the third paragraph the case has been considered in which we study the properties of a superconducting cylinder using the tunnel effect technique. We have therefore computed the dependence of the zero-bias differential conductance on the external magnetic field and on the quantum number n , for a tunnel junction of the "superconducting metal-insulator-normal metal" (S-I-N) type, in which the superconductor has a multiply connected geometry.

Some of the expressions obtained here have already been reported in the literature^(1 + 4), but in a more approximate form than we use. In the present work we have reconsidered and modified the usually accepted approximations (which in certain cases have a very small validity range) thus obtaining formulas with wider usefulness.

2. - THE GINZBURG-LANDAU EQUATIONS AND THE ORDER PARAMETER
IN THE CASE OF A SUPERCONDUCTING CYLINDER. -

The calculation of the dependence of the order parameter on the external magnetic field and on the quantum number n can be developed in a rela

tively easy manner using the theory of Ginzburg-Landau which is valid in the limit $T \sim T_c$.

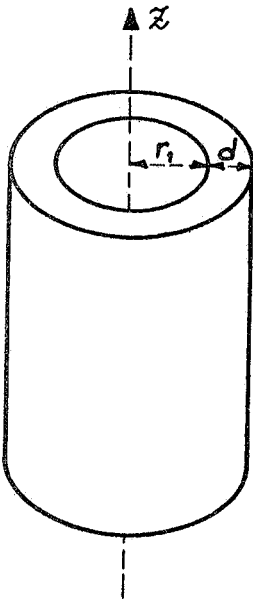


FIG. 1 - Schematic drawing of a superconducting cylinder.

Let us consider a superconducting cylinder immersed in a uniform external magnetic field parallel to the z axis. Let the internal radius be r_1 and the thickness of the wall be d (see Fig. 1). The equations of Ginzburg-Landau are written:

$$(1) \quad \left(\vec{\nabla} + \frac{ie^x}{\hbar c} \vec{A} \right)^2 \psi + \frac{k^2}{\lambda^2} (1 - |\psi|^2) \psi = 0$$

$$(2) \quad \nabla^2 \vec{A} = \frac{i\hbar c}{2e^x \lambda^2} (\psi^x \vec{\nabla} \psi - \psi \vec{\nabla} \psi^x) - \frac{|\psi|^2}{\lambda^2} \vec{A} = \frac{4\pi}{c} \vec{j}$$

with the boundary conditions given by:

$$(3) \quad (i\hbar \vec{\nabla} \psi - \frac{e^x}{c} \psi \vec{A})_{\perp} = 0$$

$$(4) \quad \vec{\nabla} \vec{A} = \vec{H}_0$$

$$(5) \quad \vec{\nabla} \vec{A} = \frac{2 \vec{A}(r_1)}{r_1}$$

where $\psi = \psi(T, \vec{A}) / \psi(T, 0)$. We recall that the penetration depth λ and the parameter K are given by the equations:

$$(6) \quad \lambda^2 = \lambda_L^2 \chi^{-1} = mc^2 / (4\pi e^x |\psi(T, 0)|^2 \chi)$$

$$(7) \quad K = K_0 \chi^{-1} = \frac{\sqrt{2} e^x \lambda^2(T) H_{cb}(T)}{\hbar c}$$

where the function χ , which gives the modification of the penetration depth due to collisions, can be approximated within 20%⁽⁵⁾ by the expression:

$$(8) \quad \chi \simeq (1 + \xi_0 / \ell)^{-1}$$

With the geometry considered, one can write:

$$(9) \quad \psi = \phi \exp(-in\Theta)$$

We will be dealing with the case in which ϕ is independent of the position which corresponds to the condition that d is much less than the coherence length $\xi_0(T)$.

Introducing some hypotheses which follow from the symmetry of the problem ($A(r) = A_\theta(r)$, $j(r) = j_\theta(r)$, $\text{div } A = 0$, $\partial / \partial z = 0$), equation (2) in cylin-

drical co-ordinates reduces to:

$$(10) \quad \frac{d}{dr} \left[\frac{1}{r} \frac{\partial}{\partial r} (rA) \right] = \frac{\phi^2}{\lambda^2} \left(A - \frac{\hbar c n}{e^x r} \right)$$

Making the substitution $A' = A - (\hbar c / e^x r) n$, equation (10) becomes a Bessel equation for A' whose integration gives:

$$(11) \quad A'(r) = C_1 I_1 \left(\frac{r\phi}{\lambda} \right) + C_2 K_1 \left(\frac{r\phi}{\lambda} \right)$$

where I and K are modified Bessel functions of the first type⁽⁶⁾. The constants of integration C_1 and C_2 are determined by the boundary conditions:

$$(12) \quad C_1 = \left[\frac{H_0 \lambda}{\phi} (K_1(\alpha) + \frac{1}{2} \alpha K_0(\alpha)) - \frac{\hbar c n}{e^x r_1} K_0(\beta) \right] / \Delta(\alpha, \beta)$$

$$C_2 = \left[\frac{H_0 \lambda}{\phi} \left(\frac{1}{2} \alpha I_0(\alpha) - I_1(\alpha) \right) - \frac{\hbar c n}{e^x r_1} I_0(\beta) \right] / \Delta(\alpha, \beta)$$

where

$$(13) \quad \Delta(\alpha, \beta) = I_0(\beta) \left[K_1(\alpha) + \frac{1}{2} \alpha K_0(\alpha) \right] + K_0(\beta) \left[I_1(\alpha) - \frac{1}{2} \alpha I_0(\alpha) \right]$$

$$(14) \quad \alpha = r_1 \phi / \lambda$$

$$(15) \quad \beta = (r_1 + d) \phi / \lambda$$

On the other hand, the magnetic field is given by the expression:

$$(16) \quad H(r) = \frac{\phi}{\lambda} \left[C_1 I_0 \left(\frac{r\phi}{\lambda} \right) - C_2 K_0 \left(\frac{r\phi}{\lambda} \right) \right]$$

A self consistent solution for ϕ can be obtained from (1). Using $\psi = \phi e^{-in\theta}$ and having $\nabla_\theta = (1/r)(\partial/\partial\theta)$, equation (1) becomes:

$$-\frac{k^2}{\lambda^2} (1 - \phi^2) \psi = \left[-\frac{n^2}{r^2} + \frac{n e^x}{\hbar c} A_0 - \left(\frac{e^x}{\hbar c} A_0 \right)^2 \right] \psi$$

This relation can be written in the more compact form:

$$\frac{k^2}{\lambda^2} (1 - \phi^2) = \left(-\frac{n}{r} + \frac{e^x}{\hbar c} A_0(r) \right)^2$$

4.

or, analogously,

$$(17) \quad \phi^2 = 1 - a^2$$

where the spatially varying terms have been properly averaged so as to satisfy the imposed condition, valid in the limit $d \ll \xi$, that ϕ is spatially constant. Consequently, a is defined as:

$$(18) \quad a^2 = \left(\frac{\lambda e^x}{k \hbar c} \right)^2 \left\langle \left(A(r) - \frac{\hbar c n}{e^x r} \right)^2 \right\rangle_{av}$$

while the average value of an operator S is defined as:

$$(19) \quad \langle S \rangle_{av} = \frac{2}{(r_1 + d)^2 - r_1^2} \int_{r_1}^{r_1 + d} r S(r) dr$$

We will not show in detail the calculations made for the explicit dependence of ϕ^2 on the external magnetic field and on n . We wish to point out, however, that in accordance with what has been mentioned in the introduction, there have been found, and then eliminated, some errors in the development of formulas reported by other researchers. Furthermore, the validity of some approximations has been re-analyzed. Therefore, our expressions will be different in some cases from those previously known. On the other hand, this permits a more consistent treatment of the correlation between the quantized paramagnetic current and the order parameter.

In the limit $d \ll \lambda$, $\lambda \ll r_1 \phi$, equation (11) becomes:

$$(20) \quad A'(r) = \frac{\hbar c}{\pi e^x r_1^2} \sqrt{\frac{r_1}{r}} \left[\frac{r_1}{2} (h_0 - n) + (r - r_1) \left(h_0 + \frac{n \gamma^2 \phi^2}{2} \right) + \frac{r_1 \phi^2}{4 \lambda^2} (r - r_1)^2 \right. \\ \left. \cdot (h_0 - n) \left[/ \left(1 + \frac{1}{2} \gamma^2 \phi^2 \right) \right] \right]$$

with

$$h_0 = \frac{e^x \pi r_1^2 H_0}{\hbar c} = \frac{\pi r_1^2 H_0}{o}$$

and

$$\gamma^2 = \frac{r_1 d}{\lambda}$$

where H_0 is the external magnetic field. With the same limitations as used in (20), the explicit expression of ϕ^2 is given by:

$$\begin{aligned}
\phi^2 = & 1 - \left(\frac{\lambda}{kr_1}\right)^2 \frac{1}{\left(1 + \frac{1}{2} \gamma^2 \phi^2\right)^2} \left\{ (h_o - n)^2 \left[\frac{1}{20} \left(\frac{d}{r_1} \gamma^2 \phi^2\right)^2 - \frac{d}{2r_1} \left(1 - \frac{2}{3} \gamma^2 \phi^2\right) + 1 \right] + \right. \\
(21) \quad & + (h_o - n) \left(h_o + n \frac{\gamma^2 \phi^2}{n}\right) \left(\frac{2d}{r_1} + \frac{d^2}{r_1^2} \frac{\gamma^2 \phi^2}{2}\right) + \frac{4}{3} \frac{d^2}{r_1^2} \left(h_o + n \frac{\gamma^2 \phi^2}{2}\right) + \\
& \left. - n \gamma^2 \phi^2 \frac{d}{r_1} \left[(h_o - n) \left(1 + \frac{1}{6} \frac{d}{r_1} \gamma^2 \phi^2\right) + \frac{d}{r_1} \left(h_o + n \frac{\gamma^2 \phi^2}{4}\right) \right] \right\} \quad (x)
\end{aligned}$$

Let us note that (21), despite the limiting conditions with which it has been deduced, can be used for the calculation of the critical field (that is, the field for which the value of the local maximum of ϕ^2 is zero). This gives the same result as in the case of the simply connected thin film. In Fig. 2 is shown a graph of ϕ^2 given by equation (21) as a function of h_o and for different values of n . The values of the parameters which appear in (21) are respectively $r_1 = 7.5 \times 10^3 \text{ \AA}$, $d = 500 \text{ \AA}$, and $\gamma^2 = 0.53$ where we have considered aluminum for which $\lambda = 2.660 \text{ \AA}$ for $T/T_c = 0.97$, $\lambda_{L(o)} = 157 \text{ \AA}$, and $\xi_o = 16.000 \text{ \AA}$. The effects of the mean free path ($l \simeq d$) on λ have been taken into account through the usual expression $\lambda \simeq \lambda_L \sqrt{1 + \xi_o/l}$.

(x) - With the further limiting conditions that $(h_o - n) \ll 1$, $(\gamma nd/r) \ll 1$, and putting $\phi^2 = 1$ on the right hand side of (21), one obtains a second approximation for ϕ^2 .

$$(22) \quad \phi^2 \simeq 1 - \left(\frac{\lambda}{kr_1}\right)^2 \left(1 + \frac{1}{2} \gamma^2\right)^{-2} \left[(h_o - n)^2 + \frac{2d}{r_1} h_o (h_o - n) + \frac{4}{3} \frac{d^2}{r_1^2} \left(h_o + \frac{n \gamma^2}{2}\right) \right]$$

Equation (22) describes a set of "parabolas" as a function of h_o and with n as a parameter. These curves intersect each other and their maxima lie on a quadratic envelop, given by the expression (23).

$$(23) \quad \phi^2 = 1 - \frac{4}{3} \left(\frac{\lambda d}{kr_1^2}\right)^2 h_o^2$$

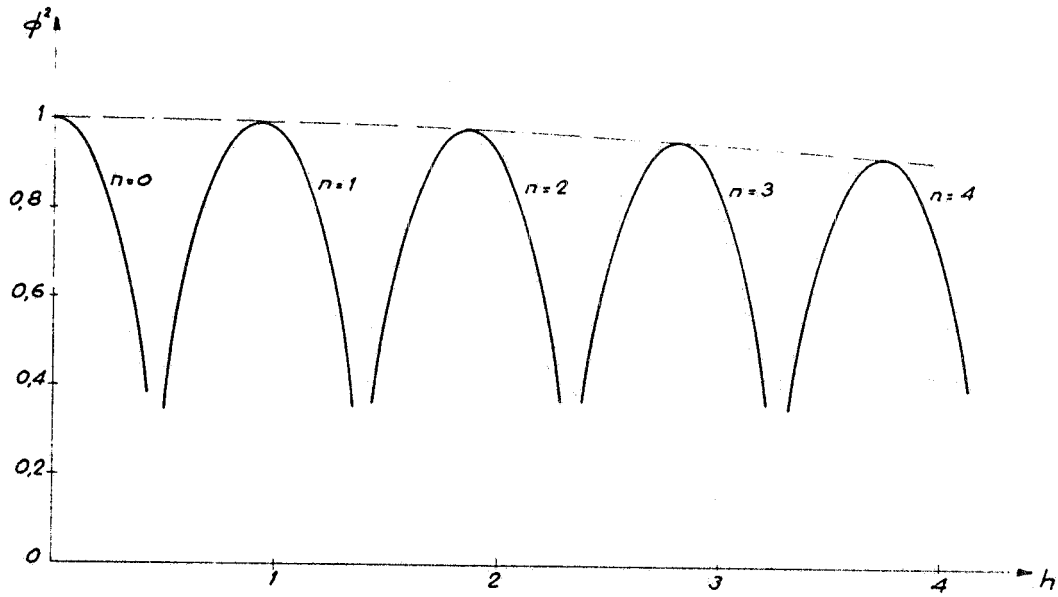


FIG. 2 - Oscillatory behaviour of the order parameter as a function of the reduced magnetic field. For the actual values of the parameters see the text.

3. - THE CORRELATION BETWEEN THE EXTERNAL MAGNETIC FIELD AND THE CURRENT DENSITY IN A HOLLOW SUPERCONDUCTING CYLINDER. -

The current density for a superconducting cylinder can be derived immediately from equation (2) (the second Ginzburg-Landau equation) once ϕ^2 is known. With some simplifying assumptions, namely $d \ll r_1$, $j(r) = j(r_1)$, the magnetic field inside the hole of the cylinder constant and equal to its value for $r = r_1$, and the flux of the magnetic field through the walls negligible with respect to the flux through the hole, one arrives at the following result:

$$(24) \quad \frac{4\pi}{c} j = \frac{1}{\pi r_1^2 d} \left(\frac{\frac{1}{2} \gamma^2 \phi^2}{1 + \frac{1}{2} \gamma^2 \phi^2} \right) \frac{h c}{e^x} (n - h_0)$$

On the other hand, if one employs this procedure and uses (21) for ϕ^2 , a physically absurd result is obtained. We find, as expected, that j is a periodic function of h_0 with the successive periodicities corresponding to various values of n . However, we also find that j shows an envelope curve increasing with applied magnetic field. The origin of this absurdity comes from the fact that the expressions (21) and (24), for ϕ^2 and j respectively, have been deduced with certain approximations. Even if these approximations don't contradict each other, they contain an unavoidable arbitrariness and therefore may influence expressions (21) and (24) in a different manner. Furthermore, we can see that the absurd result is caused analytically by the fact that in the

expressions of ϕ^2 and of j as a function of h_0 , the local maxima of ϕ^2 corresponding the various n do not lie at the same field values as the zero values of j . Consequently, we tried to eliminate some of the approximations made; in particular, we did not require that $j = j(r_1) = \text{constant}$ in the entire thickness of the cylinder. Rather, we imposed the condition that in correspondence to the values of h_0 for which ϕ^2 has a local maximum, the distribution of super currents in the cylinder wall be such as to minimize the associated kinetic energy. Without presenting all the details, let us mention that even in this case we found some physical incongruities. In fact, there is an asymmetry in the dependence of j on n such that the positive maximum values of j are smaller in absolute value than the corresponding negative ones, despite the fact that they occur at lower fields.

At this point, we changed the scheme of calculation adopting the following procedure. We first introduced a position dependent current density starting directly from the expression which expresses the quantization of the fluxoid:

$$(25) \quad \Phi = \int_S \vec{H} \cdot \vec{n} \, dS + c \oint_{C'} \mathcal{L} \vec{j} \cdot \vec{dl} = n \varphi_0$$

where $\mathcal{L} = m/n_s e \hbar^2$ and $\varphi_0 = hc/e \hbar$. From (25) it is possible to obtain for a cylindrical geometry

$$(26) \quad j(r) = \frac{1}{c \mathcal{L}} \frac{1}{2 \pi r} (n \varphi_0 - \varphi(r))$$

with

$$\varphi(r) = \int_0^r H(r') r' \, dr'$$

If we now put into (26) $\mathcal{L} = 4 \pi \lambda^2 / e^2 \phi^2$ (a relation which is derived immediately from the definition of \mathcal{L} in the Ginzburg-Landau theory where the density of superelectrons in a magnetic field is $n_s = \phi^2 / \psi(T, 0)^2$). We then obtain:

$$(27) \quad j(r) = \frac{c \phi^2}{8 \pi^2 \lambda^2 r} (n \varphi_0 - \varphi(r))$$

Obviously for (27) to be used in practice, we must know the distribution of the magnetic field on the inside and in the wall of the cylinder. Adopting then the same approximations with which we obtained (24), we find for j the following expression:

$$(28) \quad j = \frac{c \varphi_0}{8 \pi^2 \lambda^2 r_1} \frac{\phi^2}{1 + 1/2 \gamma^2 \phi^2} (n - h_0)$$

8.

Equation (28) is quite similar to (24) reported above mainly with respect to the dependence of j on ϕ^2 . It is obvious that we cannot introduce the expression (21) for ϕ^2 into (28) without running into the difficulties mentioned before.

Now it is useful to remember that problems in correlating ϕ^2 and j arose analytically from the fact that the local maxima of ϕ^2 were at values of h_0 different from those for which j was zero.

On the other hand, from a physical point of view it is reasonable, at least for small values of h_0 and n , that the correlation between ϕ^2 and j be such that the local maxima of ϕ^2 occur at the same values of h_0 for which j is locally zero.

With regard to this point, we should recall that the presence of a superconducting current different from zero increases the free energy of the superconductor towards the value which characterizes the normal state. According to this criterium, then, we have modified the expression for j simply by imposing the condition that the local maxima of ϕ^2 occur for those values of h_0 for which j is locally zero. Analytically such modification has been accomplished by transforming the factor $(n - h_0)$ that appears in (28) into $(bn - h_0)$, where b is a number near but less than 1. The analytic expression of b is the following:

$$(29) \quad b = \frac{2 + \frac{d}{r_1} + \frac{2}{3} \frac{d}{r_1} \gamma^2 + \frac{1}{6} \frac{d^2}{r_1^2} \gamma^2 + \frac{1}{60} \frac{d^2}{r_1^2} \gamma^4}{2 + \frac{3d}{r_1} + \frac{2}{3} \frac{d}{r_1} \gamma^2 + \frac{8}{3} \frac{d^2}{r_1^2} + \gamma^2 \frac{d^2}{r_1^2} + \frac{1}{10} \frac{d^2}{r_1^2} \gamma^4}$$

Using the parameters already employed in the calculation of ϕ^2 , whose graph is shown in Fig. 2, we found $b = 0.93$.

Finally the expression for j becomes:

$$(30) \quad j = \frac{c \varphi_0}{8 \pi^2 \lambda^2 r_1} \frac{\phi^2}{(1 + (1/2) \gamma^2 \phi^2)} (bn - h_0)$$

The dependence that one obtains for j as a function of h_0 and n by substituting the expression (21) for ϕ^2 into (30) is perfectly consistent from a physical point of view. Fig. 3 shows the graph of such an expression with the values of the parameters used previously.

Before concluding this paragraph, we wish to mention that, although the adopted procedure gives results which are physically consistent, it is subject to the reservation that it has been introduced "a posteriori" in the equations. But we recall that the Ginzburg-Landau equations, the starting point of all our arguments, cannot be resolved in practice without introducing suitable approximations which can easily give rise to the analytical anomalies

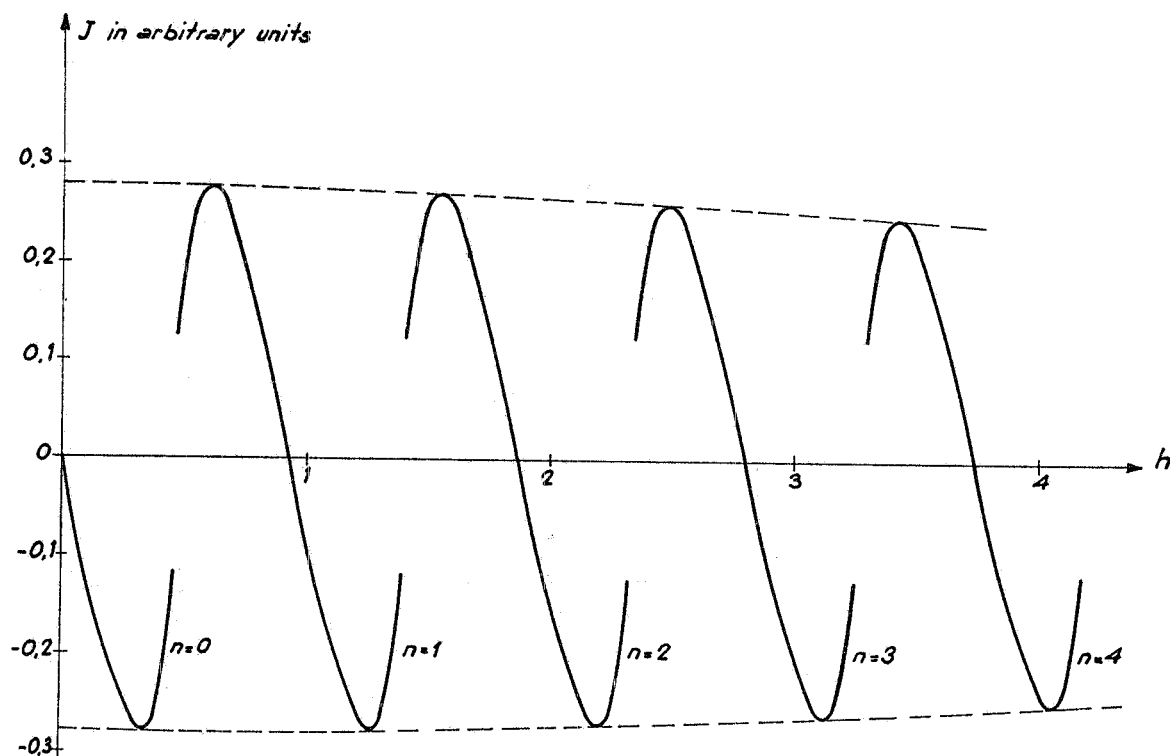


FIG. 3 - Oscillatory behaviour of the superconducting current density as a function of the reduced magnetic field.

between the expressions (21) of ϕ^2 and (25) of j .

It is worth mentioning, finally, that our assumptions are partially confirmed by the fact the critical field for the cylinder can be obtained by calculating the magnetic field for which the value of the local maximum of j is zero. This value of H_c is again the same as the one deduced from the usual expression for the critical field of thin films.

4. - THE CORRELATION BETWEEN THE MAGNETIC FIELD AND THE ZERO BIAS CONDUCTIVITY OF A TUNNEL JUNCTION FOR A SUPERCONDUCTING CYLINDER. -

We consider now the case of a tunnel junction of the S-I-N type where one of the electrodes has the form of a superconducting cylinder. Fig. 4 illustrates schematically the experimental situation we will examine. Precisely, we want to determine the dependence of the initial conductivity:

$$\sigma(0) = \left[\left(\frac{dI}{dV} \right)_{NS} / \left(\frac{dI}{dV} \right)_{NN} \right]_{V=0}$$

of a tunnel junction on the external magnetic field, taking into account the effects introduced by the quantization of the fluxoid due to the multiply connected geometry.

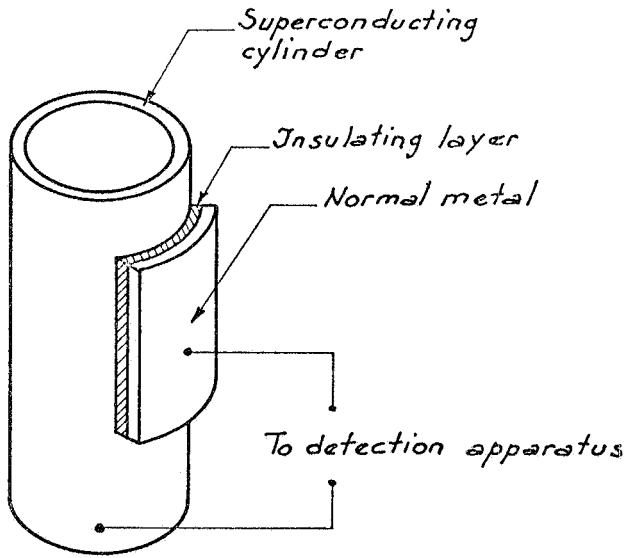


FIG. 4 - Schematic drawing of a tunnel junction suitable for an experiment with a multiply connected superconductor.

can be described by a parameter Z that, in our case, is linked implicitly to the supercurrent by the relation:

$$(31) \quad j/j_{\max} = 0.947 Z^{3/4} \left(\pi - \frac{4}{3} Z^{3/2} \right) \exp \left(- \frac{3}{8} \pi Z^{3/2} \right)$$

valid in the limit $Z < 1$ ⁽⁸⁾. Also, the structure of the theory is such that when there are several depairing factors present in the superconductor, their total effect is taken into account by introducing a parameter Z_{tot} which is equal to the sum of the single parameters due to the various depairing agents. Returning now to the calculation of $\sigma(0)$ as a function of H for the case of a superconducting cylinder, it is reasonable to think that there are two depairing factors present: one due to the persistent paramagnetic current associated with the successive values of n , and the other due to the external magnetic field. We notice that the expression (30) for j represents the total current density (paramagnetic + diamagnetic), and so it is perfectly consistent that this current goes periodically to zero with increasing h . It is practically impossible to extract from (30) the only paramagnetic contribution since ϕ^2 , too, depends both on n and on h . As a reasonable approximation, which is certainly valid for low fields and for very thin wall thickness (which means we can neglect the diamagnetic contribution), we have assumed that (30) describes the only paramagnetic current. The parameter Z_{tot} then becomes:

$$(32) \quad Z_{\text{tot}} = Z_j + Z_h$$

where Z_j is given by an expression analogous to (31) in which j_{\max} assumes

First we recall how $\sigma(0)$ depends on the current flowing in the superconducting part of a tunnel junction in the case of a simply connected geometry⁽⁷⁾. As is known, the effect of such a current can be introduced by treating the current as one of the many depairing factors which can exist in a superconductor. The results of the theory are not expressible in analytically simple terms (for more details on this point see ref.(7), but it is possible to compute numerically the dependence of $\sigma(0)$ on the ratio j/j_{\max} . Such a dependence reflects the modifications induced on the state density and on the excitation gap by the superconducting current. Furthermore, we recall that the presence of the depairing factor

successively the values of the local maxima of j corresponding to various n , while Z_h takes into account the depairing action of the external magnetic field and is given by the expression⁽⁹⁾:

$$(33) \quad h/h_c = \sqrt{2} Z_h^{3/4} \exp\left(-\frac{1}{8} \pi Z_h^{3/2}\right)$$

which is also valid for $Z_h < 1$.

We want to point out that the situation we are considering is rather unusual in the study of depairing phenomena, for we have the superposition of two factors: one that increases in a monotonic manner, as well as one that oscillates and periodically goes to zero. This behaviour of the depairing factors is peculiar to the problem we are treating and is a consequence of the multiply connected geometry.

Inserting now the parameter (32) into the various equations that appear in the depairing theory and following the same procedure used in other cases, we have computed numerically the dependence of $\sigma(0)$ on h , thus obtaining the graph illustrated in Fig. 5. As one can see, $\sigma(0)$ has an oscillatory dependence with successive minima lying on an increasing envelope that reaches the value $\sigma(0) = 1$ (corresponding to the situation when the junction is completely in the normal state) just when $h = h_c$.

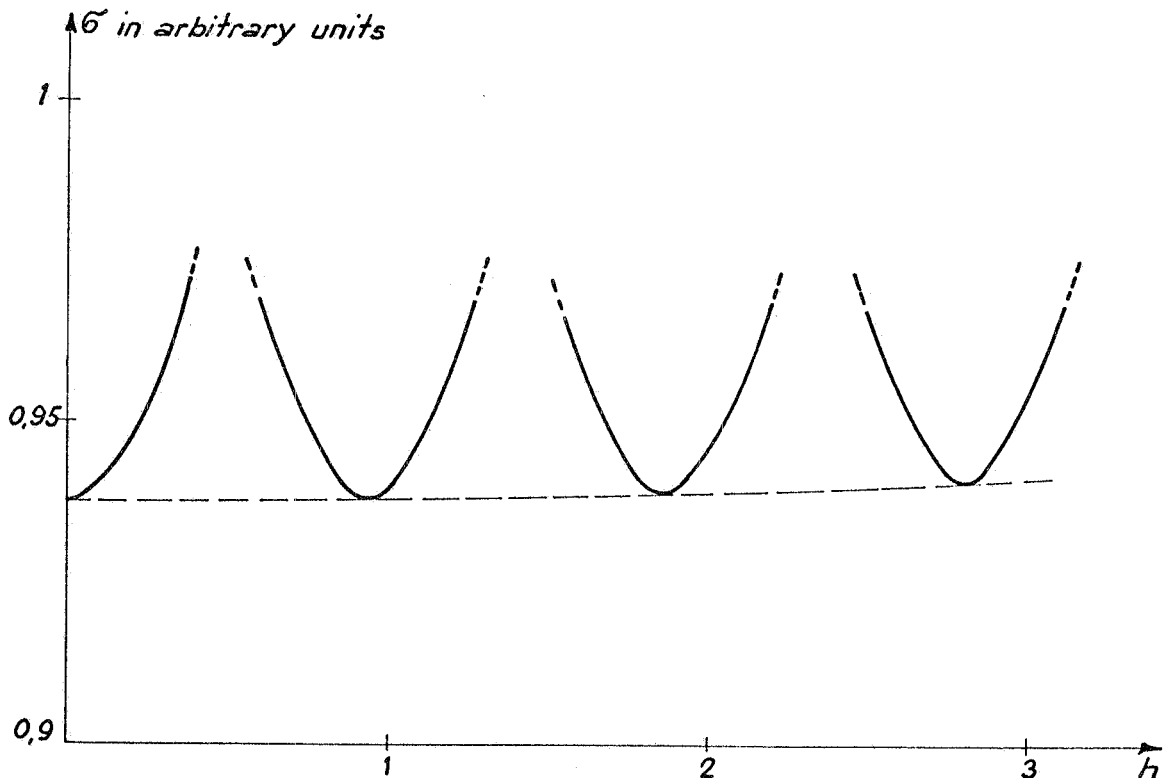


FIG. 5 - Oscillatory behaviour of the zero-bias conductivity as a function of the reduced magnetic field for the situation shown in Fig. 4.

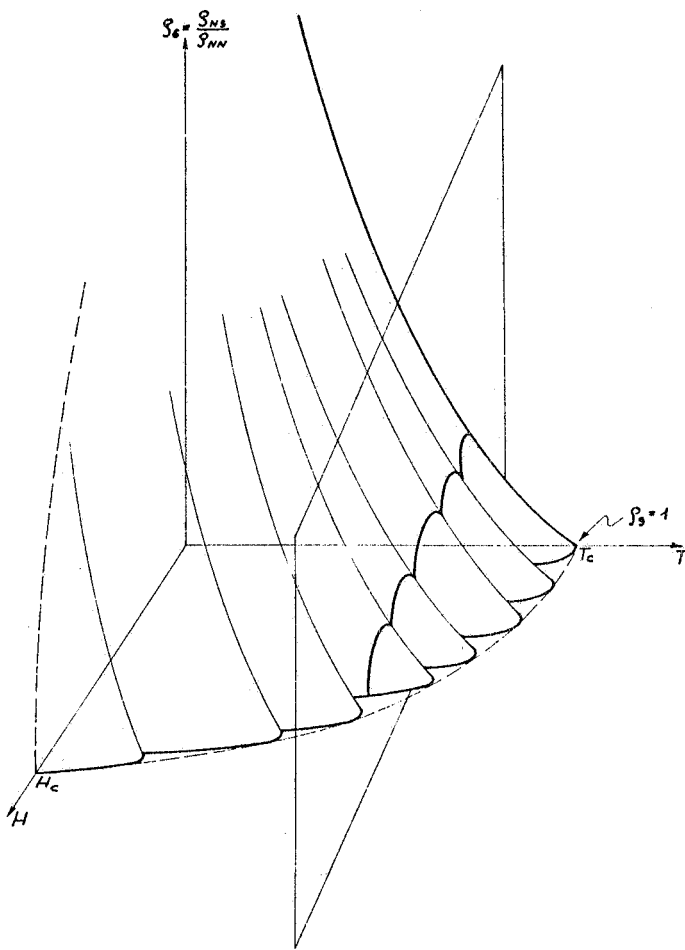


FIG. 6 - Qualitative three-dimensional representation of the zero-bias tunnel resistivity as a function of the temperature and magnetic field.

It is possible at this point to summarize graphically our results by making a three dimensional representation of the following quantities: temperature, applied magnetic field, and initial tunnel resistance for a S-I-N junction (Fig. 6). Clearly in the H, T plane one finds the well-known curve showing the periodic dependence of T_c on H_c . Furthermore, if one considers the intersection of an isothermal plane (that is, a plane parallel to the H, R_g axes), for a temperature near T_c , with the three dimensional surface, one observes the oscillatory dependence of R_g on H which is practically the same as that reported in Fig. 5. As T decreases, the separation between the various periodicities tends to disappear, mainly due to the fact that the parameter γ^2 , depending on the temperature through λ , increases to values greater than 1. Therefore (see ref. 1) this implies that all of the oscillatory parts of the various quantities we have examined (ϕ^2 , j , and R_g) gradually overlap.

REFERENCES. -

- (1) - D. H. Douglass Jr., Phys. Rev. 132, 513 (1963).
- (2) - R. D. Parks and W. A. Little, Phys. Rev. 133, A97 (1964).
- (3) - R. P. Graff and R. D. Parks, Phys. Rev. 176, 567 (1968).
- (4) - L. Meyers and R. Meservey, Phys. Rev. B4, 824 (1971).
- (5) - M. Tinkham, Phys. Rev. 110, 26 (1958).
- (6) - A. Erdelyi, W. Magnus, F. Oberhettinger and F. Tricomi, Higher Transcendental Functions (McGraw-Hill, New York, 1953), vol. 2, p. 9.
- (7) - G. Paternò, M. V. Ricci and N. Sacchetti, Phys. Rev. B3, 3792 (1971).
- (8) - P. Fulde, Phys. Rev. 137, A783 (1965).
- (9) - J. L. Levine, Phys. Rev. 155, 373 (1967).