

cipally to the fact that Klein's analysis (which underestimates the role of states with  $L \geq 2$ ) agrees badly with the experimental data on  $n$ - $p$  scattering in the angular region close to  $180^\circ$ .

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## On the Magnetic Properties of Superconductors of the Second Group

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A study is made of the magnetic properties of bulk superconductors for which the parameter  $\kappa$  of the Ginzburg-Landau theory is greater than  $1/\sqrt{2}$  (superconductors of the second group). The results explain some of the experimental data on the behavior of superconductive alloys in a magnetic field.

THE AUTHOR<sup>1</sup> has already noted that the quasi-microscopic Ginzburg-Landau theory<sup>2</sup> of superconductivity leads to the conclusion that there exist two groups of superconductors. For the first of these groups, the parameter  $\kappa$  entering into the Ginzburg-Landau theory is less than  $1/\sqrt{2}$ , and for the second group it is greater than  $1/\sqrt{2}$ .

This parameter  $\kappa$  determines to a great extent the surface energy at the normal - superconducting interface. It has already been mentioned,<sup>2</sup> in particular, that the calculated surface energy of a superconductor with  $\kappa > 1/\sqrt{2}$  is negative. Thus superconductors of the second group should have properties very different from those of the first group.

For pure metal,  $\kappa$  is found to be small. For instance for mercury,  $\kappa = 0.16$ . In view of this,

Ginzburg and Landau considered only the case in which  $\kappa \ll 1/\sqrt{2}$ .

It has been shown by Zavaritskii,<sup>3</sup> however, that the properties of pure metal thin films condensed at liquid-helium temperatures are not described by such a theory. Zavaritskii and the present author have therefore suggested that such films correspond to  $\kappa > 1/\sqrt{2}$ , and that superconductors can thus be divided into two groups. The critical field for superconductors of the second group has already been calculated<sup>1</sup> as a function of the film thickness. The agreement obtained with Zavaritskii's experimental data was not bad.

In the present work, a more detailed investigation of the magnetic properties of bulk superconductors of the second group (a cylinder in a longitudinal field) is undertaken. The results obtained show

that  $\kappa > 1/\sqrt{2}$  for a large number of superconducting alloys whose magnetic properties had not previously been well understood.\*

### 1. TRANSITION TO THE NORMAL STATE

It was shown by Ginzburg and Landau<sup>2</sup> that if  $\kappa > 1/\sqrt{2}$ , the superconductivity is maintained at fields greater than  $H_{cm}$  at which equilibrium could exist between the normal and superconducting states. At fields higher than  $H_{cm}$ , a state with  $\Psi = 0$  is unstable, and superconducting sections with  $\Psi \neq 0$  may arise. It was shown<sup>1</sup> that this instability continues to some value  $H_{c2}$ , which for a bulk superconductor is  $\kappa\sqrt{2}H_{cm}$ . At this value of the field the superconductor undergoes transition to the normal state by means of a second-order phase transition. In this section we shall investigate the properties of a superconductor in the neighborhood of the transition point, that is

$$\kappa - H_0 / \sqrt{2}H_{cm} \ll \kappa. \quad (1)$$

The Ginzburg-Landau equations (in units previously used by the author,<sup>2</sup> and to be used henceforth),† can be written

$$\left(\frac{i\nabla}{\kappa} + \mathbf{A}\right)^2 \Psi = \Psi - \Psi^* |\Psi|^2, \quad (2)$$

$$-\text{curl curl } \mathbf{A} = |\Psi|^2 \mathbf{A} + \frac{i}{2\kappa} (\Psi^* \nabla \Psi - \Psi \nabla \Psi^*). \quad (3)$$

We shall assume that the superconductor fills all space, and that the external field  $H_0$  is directed along the  $z$  axis. Let the potential  $A$  be directed along the  $y$  axis.

Close to the transition point  $|\Psi|^2 \ll 1$ , so that in the first approximation we may neglect the influence of  $\Psi$  on the field. We then obtain

$$H = \text{const} = H_0, \quad A = H_0 x. \quad (4)$$

We note that the point  $x = 0$  may be located anywhere in space. Let us now go on to Eq. (2). Inserting (4) into the equation for  $\Psi$ , neglecting the term  $|\Psi|^2 \Psi$  and considering  $\Psi$  to be a function of  $x$  only in the first approximation, we obtain the oscillator type of equation

$$d^2 \Psi / dx^2 - \kappa^2 (1 - H_0^2 x^2) \Psi(x) = 0, \quad (5)$$

as found previously.<sup>2</sup> This equation has solutions when  $H_0 = \kappa/(2n + 1)$ . In particular, the largest value  $H_0 = \kappa$  corresponds to

$$\Psi = \exp \{-\kappa^2 x^2 / 2\}. \quad (6)$$

In addition to such solutions, Eq. (2) is satisfied by the function

$$\exp \left\{ iky - \frac{\kappa^2}{2} \left( x - \frac{k}{\kappa^2} \right)^2 \right\}. \quad (7)$$

Since the point  $x = 0$  has no special properties, the conditions will be exactly the same at all points of space, and it is natural to pick  $\Psi$  in the form

$$\Psi = \sum_{n=-\infty}^{\infty} C_n e^{ikny} \psi_n(x), \quad (8)$$

$$\psi_n(x) = \exp \left[ -\frac{\kappa^2}{2} \left( x - \frac{kn}{\kappa^2} \right)^2 \right]$$

with arbitrary coefficients  $k$  and  $C_n$ .

This form of  $\Psi$  is a solution of the linear equation, and refers in fact to  $H_0 = \kappa$ . In order to find a solution for  $H_0 < \kappa$ , let us take into account the nonlinear terms in the equations. We find the approximation of interest by inserting  $A$  from Eq. (4) with  $H_0 = \kappa$ , and  $\Psi$  from Eq. (8) into Eq. (3). This leads to

$$\begin{aligned} & \frac{\partial^2 A}{\partial x^2} \\ &= \sum_{n,m} C_n C_m^* e^{i(n-m)ky} \left[ \kappa k - \frac{(n+m)k}{2\kappa} \right] \psi_n(x) \psi_m(x), \quad (9) \\ & - \frac{\partial^2 A}{\partial x \partial y} = \frac{ik}{2\kappa} \sum_{n,m} C_n C_m^* e^{i(n-m)ky} (n-m) \psi_n(x) \psi_m(x). \end{aligned}$$

It can be shown easily that

$$H = \partial A / \partial x = H_0 - |\Psi|^2 / 2\kappa; \quad (10)$$

$$A = H_0 x - \frac{1}{2\kappa} \int |\Psi|^2 dx, \quad (11)$$

is a solution of this equation, where  $\Psi$  is given by Eq. (8), and  $H_0$  is some still undetermined constant. It will be shown later that this constant is equal to the external field strength.

Let us now consider Eq. (2) in the following approximation. We insert into it Eq. (11), and take account of the term  $\Psi |\Psi|^2$ . We shall assume in this approximation that the function is still of the form of Eq. (8), but we shall add small terms  $\psi_n^{(1)}(x)$  to

\* The suggestion that  $\kappa$  may be greater than  $1/\sqrt{2}$  for alloys was first made by L. D. Landau.

† We note that in these units  $H_{cm} = 1/\sqrt{2}$ .

all the  $C_n \psi_n(x)$ . The equations for these small terms are of the form

$$\begin{aligned} & \left(\frac{kn}{x} - \kappa x\right)^2 \psi_n^{(1)} - \frac{1}{x^2} \frac{d^2 \psi_n^{(1)}}{dx^2} - \psi_n^{(1)} = 2(x - H_0) \kappa \left(x - \frac{nk}{x^2}\right) C_n \psi_n(x) + \\ & + \sum_{p, m} C_p C_m^* C_{n-p+m} \left\{ \left[ x - \frac{k}{x^2} \left( n - \frac{p-m}{2} \right) \right] \psi_{n-p+m}(x) \int_0^x \psi_p(x') \psi_m(x') dx' - \psi_p(x) \psi_m(x) \psi_{n-p+m}(x) \right\}. \end{aligned} \quad (12)$$

Thus  $\psi_n^{(1)}$  is a solution of an inhomogeneous equation. But in order for such a solution to exist, it is necessary that the right side of the equation be orthogonal to the solution of the corresponding homogeneous equation. The solution of the homogeneous equation is just the function  $\psi_n(x)$ . Performing the necessary integration, we obtain the following condition:

$$\left(\frac{1}{2x^2} - 1\right) \sum_{p, m} C_p C_m^* C_{n-p+m} \exp\left\{-\frac{k^2}{2x^2} [(p-n)^2 + (m-n)^2]\right\} + \frac{x-H_0}{x} C_n = 0. \quad (13)$$

Let us multiply this equation by  $C_n^*$  and sum over  $n$ . It is easily verified that we then obtain

$$\frac{x-H_0}{x} \overline{|\Psi|^2} + \left(\frac{1}{2x^2} - 1\right) \overline{|\Psi|^4} = 0, \quad (14)$$

where  $\Psi$  is the function of Eq. (8). Thus  $H_0$  enters into  $\Psi$  in the form of a coefficient.

The quantity  $H_0$  can be expressed in terms of the magnetic induction. According to (10) and (14), we have

$$B = \overline{H} = H_0 - \overline{|\Psi|^2} / 2\kappa = H_0 - (x - H_0) / (2x^2 - 1)\beta, \quad (15)$$

where

$$\beta = \overline{|\Psi|^4} / (\overline{|\Psi|^2})^2 \quad (16)$$

is independent of  $H_0$ .

Let us now calculate the free energy. According to Ginzburg and Landau,<sup>2</sup> the free energy is

$$\frac{F_{sH} - F_{s0}}{H_{cm}^2 / 4\pi} = \int dV \left\{ \frac{1}{2} - |\Psi|^2 + \frac{|\Psi|^4}{2} + H^2 + \left( -\frac{i\nabla\Psi}{x} - \mathbf{A}\Psi \right) \left( \frac{i\nabla\Psi^*}{x} - \mathbf{A}\Psi^* \right) \right\}. \quad (17)$$

If we now make use of (2) and (3) and neglect the surface integral, which is of no importance to us, we obtain

$$\frac{F_{sH} - F_{s0}}{H_{cm}^2 / 4\pi} = \int \left( H^2 - \frac{|\Psi|^4}{2} + \frac{1}{2} \right) dV. \quad (18)$$

The free energy per unit volume is proportional to the average of the integrand. Denoting this average by  $F_1$  and writing it in terms of  $B$ , we obtain

$$F_1 = \frac{1}{2} + B^2 - \frac{(x-B)^2}{1+(2x^2-1)\beta}. \quad (19)$$

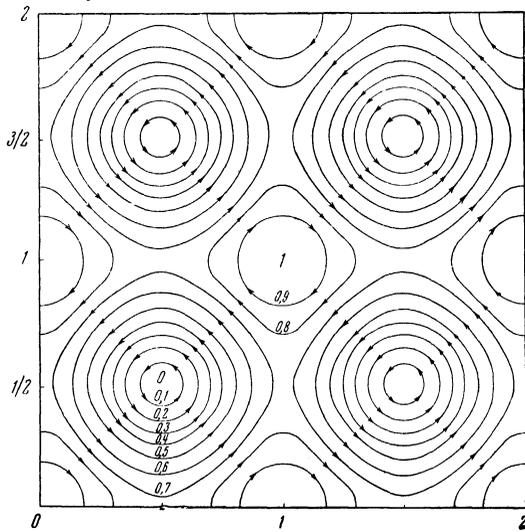
For a given  $B$  this quantity will be minimum when  $\beta$  has its lowest value.

We note that (13) can be written in the form

$$\partial\beta / \partial C_n^* = 0, \quad (13')$$

becoming equivalent to the condition that  $\beta$  is a minimum. It is seen from (16) that  $\beta$  cannot be less than unity.

The condition that  $\Psi$  be periodic in  $x$  is the following simple recursion relation for the  $C_n$ :



$$C_{n+N} = C_n. \quad (20)$$

The general analysis of the problem of choosing the coefficients is extremely complicated, but there is no reason to suppose that the actual value of  $N$  is a very large number. It would seem that the lowest energy corresponds to the simplest case\*  $N = 1$ . In this case  $\beta$  is given by

$$\beta = \frac{k}{x\sqrt{2\pi}} \left( \sum_n \exp \left\{ -\frac{k^2}{2x^2} n^2 \right\} \right)^2.$$

The minimum value of  $\beta$  is obtained when  $k = \kappa\sqrt{2\pi}$ , and is equal to

$$\psi = \left( \sum_n e^{-\pi n^2} \right)^2 = \vartheta_3^2(0, 1) = 1.18. \quad (21)$$

Then  $\Psi$  is

$$\Psi = Ce^{-\kappa^2 x^2} \vartheta_3[\sqrt{2\pi} \kappa i(x + iy); 1]. \quad (22)$$

It can be shown from the properties of the function  $\vartheta_3$  that rotation by  $90^\circ$  multiplies  $\Psi$  by the phase factor  $e^{i\kappa^2 xy}$ , but does not change it otherwise.

\* In addition to this case, which will be seen later to lead to a square lattice, we considered the example of a triangular lattice. The side of the triangle is  $(1/\kappa)\sqrt{8\pi/3\sqrt{3}}$ . By numerical integration, however, we were able to show that this lattice corresponds to a larger value of  $\beta$ , namely  $\beta = 1.32$  (see also the next section).

Thus  $|\Psi|^2$  has the symmetry of a square lattice. When  $x = y = (1/\kappa)\sqrt{\pi/2}$ , the function  $\Psi$  vanishes. According to (10),  $H$  attains its maximum value  $H_0$  at these points.

We shall now show that  $H_0$  is equal to the field strength. As is well known, the field strength is  $4\pi\partial F/\partial B$  (in the usual units). This leads to a relation between  $H$  and  $B$  which is identical with Eq. (15). In order to perform the comparison with experiment (this will be done in Sec. 3) more conveniently, the latter can be rewritten in terms of the function  $M(H_0)$ ,

$$-4\pi M/(H_{c2} - H_0) = 1/1.18(2x^2 - 1), \quad (23)$$

where  $M$  and  $H$  can be taken in the usual units. We note that the magnetic moment is proportional to the first power of  $H_{c2} - H_0$ .

Equation (23) is the macroscopic characterization of the behavior of a bulk sample in an external field. As for the microscopic structure of the field penetration, collecting the results described above we obtain the following picture. In a plane perpendicular to the field direction, the structure is periodic with the symmetry of a square lattice. The field varies between its maximum value  $H_0$  and its minimum value given by

$$H_{\min} = H_0 - (H_{c2} - H_0)\sqrt{2}/(2x^2 - 1). \quad (24)$$

When the applied field strength  $H_0$  is increased, the field between the maxima increases up to values equal to  $H = H_0 = \kappa$ .

The field and current distribution is shown in the figure. The lines correspond to  $|\Psi|^2 = \text{const}$  (normalized to  $|\Psi|_{\max}^2 = 1$ ) or equivalently, to  $H = \text{const}$ . These are then the streamlines (their directions being given by the arrow).

## 2. THE BEGINNING OF FIELD PENETRATION INTO A SUPERCONDUCTOR

In the preceding section we concluded that in the neighborhood of the transition point to the normal state, superconductors of the second group are in a special state which is neither normal nor superconducting, and differs significantly from the intermediate state which may arise in noncylindrical bulk samples. Let us call this the mixed state. One naturally wonders how this state changes as the field is decreased, and where and how the transition to superconductivity occurs.

If we direct our attention to the experimental

$B(H_0)$  curves for alloys,<sup>4</sup> we see that this transition can be both a first and a second order phase transition. It would seem (see Sec. 3) that the first order phase transition occurs when  $\kappa$  is not very large, whereas the second order transition occurs when  $\kappa \gg 1$ . It is extremely difficult to explain this on theoretical grounds, since this would require solving Eqs. (2) and (3) without any simplifying assumptions.

Let us consider only the simplest case of a second order phase transition, which would seem, as has already been mentioned, to take place when  $\kappa$  is not too small.

Let us start with a determination of the transition point. It is natural to assume that when the field first begins to penetrate, the structure of this penetration is again of the lattice type, although with a very large period. At the lattice points the field will have its maximum value, and will practically vanish between them. Thus the field penetration pattern is that of separated filaments. This pattern reminds one of the distribution of superfluidity in helium II by the creation of vortex filaments, as proposed by Onsager<sup>5</sup> and Feynman.<sup>6</sup> The similarity is even greater when one analyzes the behavior of the  $\Psi$  function in the neighborhood of these filaments in greater detail. When one considers the situation at fields close to  $H_{c2}$ , one sees easily that in the neighborhood of those points where  $\Psi$  vanishes and the field is maximum, the phase of the  $\Psi$  function is\*  $\theta$ , where  $\theta$  is the polar angle, and thus changes by  $2\pi$  in going once around such a point. It is natural to suppose that the phase behaves in the same way in the neighborhood of separate filaments when the separation between them is large. As will be seen later, the  $\Psi$  function vanishes also in the center of a filament. This behavior of the  $\Psi$  function, in particular of its phase, has been suggested by Feynman in application to vortices in helium II.

When the filaments are sufficiently separated, they can be considered independent of each other, and each filament can be treated separately. In this case, the problem has cylindrical symmetry. Let us introduce the modulus and phase of the  $\Psi$  function

$$\Psi = f e^{i\varphi} \quad (25)$$

\* In the neighborhood of such a point with coordinates  $(x_1, y_1)$ ,  $\Psi$  is proportional to  $(x - x_1) + i(y - y_1)$ .

and consider the vector  $\mathbf{A}$  directed perpendicular to the radius vector. We introduce the scalar quantity  $Q$ , which is equal to the absolute value of the vector  $\mathbf{A} - \nabla\varphi/\kappa$ . Equations (2) and (3) then become

$$-\frac{1}{\kappa^2 r} \frac{d}{dr} \left( r \frac{df}{dr} \right) + Q^2 f = f - f^3, \quad (26)$$

$$\frac{d}{dr} \left[ \frac{1}{r} \frac{d}{dr} (rQ) \right] = Qf^2. \quad (27)$$

The field  $H$  is given by

$$H = -\frac{1}{r} \frac{d}{dr} (rQ). \quad (28)$$

The statement of the problem requires that  $f \rightarrow 1$ ,  $H \rightarrow 0$ ,  $Q \rightarrow 0$  as  $r \rightarrow \infty$ . Further, we have assumed that for a separate filament  $\varphi = \theta$ . But this leads to  $\nabla_{\theta}\varphi = \partial\varphi/r\partial\theta = 1/r$ . In other words,  $Q \rightarrow 1/\kappa r$  as  $r \rightarrow 0$ . According to the theory of Onsager and Feynman the superfluid velocity  $v_s$  in helium II diminishes in the same way, with increasing distance from the center of the filament.

In addition, it is of course necessary to require that  $f$  be everywhere bounded. These conditions uniquely determine the solution of (26) and (27). In general, however, this solution can be obtained only by numerical integration, and this must be performed separately for each value of  $\kappa$ .

When  $\kappa \gg 1$ , the solution is considerably simplified. We note for this purpose that the distance over which  $Q$  should have a large variation is  $r \sim 1$ . As for  $f$ , it varies to a large extent at distances  $r \sim 1/\kappa$ . In view of this, one may assume that  $f$  has already attained the value  $f = 1$  in Eq. (26), from which we immediately obtain

$$Q = K_1(r)/\kappa, \quad (29)$$

where  $K_1$  is the Hankel function of imaginary argument. Further, we note that  $Q = 1/\kappa r$  when  $r \ll 1$ . Inserting this into the equation for  $f$  we obtain

$$\frac{1}{\kappa^2} \left[ \frac{1}{r} \frac{d}{dr} \left( r \frac{df}{dr} \right) - \frac{1}{r^2} f \right] = f^3 - f. \quad (30)$$

At distances  $r \gg 1/\kappa$ , the solution of this equation is  $f^2 = 1 - 1/\kappa^2 r^2$ , and when  $r \ll 1/\kappa$ , the solution is  $f = Cr$ , where  $C$  is a constant which can be obtained by numerical solution of (30).

Let us now calculate  $\varepsilon$ , the free energy per unit

length of a filament. To do this we must evaluate the integral

$$\begin{aligned} \varepsilon &= 2\pi \int_0^{\infty} \left( H^2 + \frac{1-f^4}{2} \right) r dr \\ &= \pi \int_0^{\infty} \left[ (1-f^4) r - f^2 \frac{d}{dr} (r^2 Q^2) \right] dr. \end{aligned} \quad (31)$$

If we note that the integrand decays exponentially when  $r \gg 1$ , and assume that  $\ln \kappa \gg 1$ , it is easily seen that the important contribution to the integral will be from the interval  $1/\kappa \ll r \ll 1$ , and we then obtain

$$\varepsilon = (2\pi/\kappa^2) \ln \kappa.$$

Numerical integration of (30) and (31) makes it possible to obtain a correction to  $\ln \kappa$ , so that we finally obtain

$$\varepsilon = (2\pi/\kappa^2) (\ln \kappa + 0.081). \quad (32)$$

In order that the formation of filaments be energetically possible, the field strength  $H_0$  must be such that  $F_1 - 2H_0 B$  be negative. If the filament density, *i.e.*, the number of filaments crossing a unit area, be denoted by  $n$ , we have

$$F_1 = n\varepsilon, \quad B = \bar{H} = n \int H dS = n \oint \mathbf{A} \cdot d\mathbf{l}. \quad (33)$$

In view of the fact that the contour of integration extends to  $r = \infty$ , and that at these large distances  $Q = 0$ , we conclude that  $\mathbf{A} = \nabla\varphi/\kappa$ , which means that its absolute value is  $1/\kappa r$ . Thus

$$B = 2\pi n / \kappa. \quad (34)$$

It follows from this that the filaments start being formed when the field strength  $H_0$  attains the value

$$H_{c1} = \varepsilon \kappa / 4\pi. \quad (35)$$

This formula is valid for arbitrary  $\kappa$  on the assumption that the phase transition is of second order. When  $\kappa \gg 1$ , we obtain

$$H_{c1} = (1/2\kappa) (\ln \kappa + 0.08). \quad (36)$$

It is interesting to note that at the center of the filament the field is about twice  $H_{c1}$  in the given case. Indeed, according to Eq. (27) we have

$$H(0) = \int_0^{\infty} Q f^2 dr = (\ln \kappa - 0.18) / \kappa. \quad (37)$$

From (35) it is seen that  $H_{c1}$  is independent of  $n$ , and that an arbitrary number of filaments may be formed in this approximation for  $H_0 > H_{c1}$ . In order to find the function  $n(H_0)$ , and therefore  $B(H_0)$ , we must take account of the interaction between the filaments.

This is easily done if  $\kappa \gg 1$ . Indeed, in this case the region in which  $f$  differs from unity ( $r \sim 1/\kappa$ ) is small compared with the distances at which the interaction between the filaments takes place ( $r \sim 1$ ). In view of this, the filament centers will play the role of singularities in the solution, and their detailed structure will not be of importance. It is not difficult to see that the equation for the field together with the conditions at the centers of the filaments can be written in the form

$$\Delta H - H = - (2\pi/\kappa) \sum_m \delta(\mathbf{r} - \mathbf{r}_m), \quad (38)$$

where the  $\mathbf{r}_m$  are the coordinates of the centers of the filaments. The solution of this equation is

$$H = \frac{1}{\kappa} \sum_m K_0(|\mathbf{r} - \mathbf{r}_m|). \quad (39)$$

The energy can be written in the form

$$\begin{aligned} F_1 &= \overline{H^2 + (\nabla H)^2} = \overline{H(H - \Delta H)} \\ &= \frac{2\pi n}{\kappa^2} \sum_m K_0(|\mathbf{r}_m|), \end{aligned} \quad (40)$$

where the  $\mathbf{r}_m$  are calculated from one of the filament centers. The term corresponding to  $\mathbf{r}_m = 0$  should be replaced by Eq. (32). The quantity  $n$  and the periodicity of the lattice, which enter into (40), can be expressed in terms of the magnetic induction  $B$  by Eq. (34). For a square lattice we obtain

$$F_1 = \frac{\kappa B}{2\pi} \varepsilon + \frac{B}{\kappa} \sum_{l^2+m^2 \geq 1} K_0 \left( \sqrt{\frac{2\pi(l^2+m^2)}{\kappa B}} \right). \quad (41)$$

It is, however, possible that the lattice is actually not a square one or that its symmetry changes as the field is varied. Therefore the problem should be dealt with more carefully. The most

probable structure which one can assume instead of the square one is the triangular one. For such a lattice one easily obtains

$$F_1 = \frac{\kappa B}{2\pi} \varepsilon + \frac{B}{\kappa} \sum_{l^2+m^2+lm \geq 1} K_0 \left( \sqrt{\frac{4\pi(l^2+m^2+lm)}{V^3 \kappa B}} \right). \quad (42)$$

By comparing the asymptotic form of this expression for small  $B$  with asymptotic form of (41),

$$F_{1\Delta} \approx \frac{\kappa B}{2\pi} \varepsilon + 6 \frac{B}{\kappa} \sqrt{\frac{\pi}{2}} \left( \frac{V^3 \kappa B}{4\pi} \right)^{1/4} \exp \left( -\sqrt{\frac{4\pi}{V^3 \kappa B}} \right), \quad (43)$$

$$F_{1\Box} \approx \frac{\kappa B}{2\pi} \varepsilon + 4 \frac{B}{\kappa} \sqrt{\frac{\pi}{2}} \left( \frac{\kappa B}{2\pi} \right)^{1/4} \exp \left( -\sqrt{\frac{2\pi}{\kappa B}} \right),$$

one sees clearly that for sufficiently small  $B$  there should be a transition to the triangular modification. The value of  $H_0$  at the transition point is determined by setting the free energy for a given field strength equal to  $F_1 - 2H_0B$ , and is

$$H'_1 = H_{c1} + 0.0394/\kappa. \quad (44)$$

This transition is clearly a first order phase transition. The induction undergoes a jump from  $B_1 = 0.286/\kappa$  to  $B_2 = 0.294/\kappa$ . At fields greater than  $H'_1$ , the square lattice is more favorable, which justifies to some degree the assumption made in the previous section.

We note that the transition takes place in the immediate neighborhood of  $H_{c1}$ , and that the discontinuity in the induction is so small (about 3%) that it would be extremely difficult to observe it on a  $B(H_0)$  curve.

It is not difficult to see that when  $H = H_{c1}$ , the  $B(H_0)$  curve has a vertical tangent. Indeed, when  $H \approx H_{c1}$ , the  $H_0$ -dependence of  $B$  is determined primarily by the term  $H_0 \sim \exp[-(4\pi/\sqrt{3}\kappa B)^{1/2}]$ , and  $dH_0/dB \rightarrow 0$  as  $B \rightarrow 0$ . In the other limiting case, when  $\kappa B \gg 1$ , the various terms of the sum of Eq. (41) have arguments which lie close to each other, so that the sum can be replaced by an integral. This gives  $F_1 \approx B^2$ , and  $H_0 \approx B$ . Thus as  $H_0$  is increased, the curve asymptotically approaches the straight line  $B = H_0$ . This goes on until the distance between the filaments becomes of the order of  $1/\kappa$ , i.e., until  $H_0 \sim \kappa$ .

The dependence of  $\kappa B$  on  $(H_0 - H_{c1})$  is given by the universal function

$$\kappa(H_0 - H_{c1}) = \frac{1}{4} \sum_{x_{l,m} \neq 0} [2K_0(x_{l,m}) + x_{l,m}K_1(x_{l,m})],$$

$$\text{where } x_{l,m} = \sqrt{4\pi(l^2 + m^2 + lm)} / \sqrt{3\kappa B}$$

$$\text{for } \kappa(H_0 - H_{c1}) < 0.0394,$$

$$x_{l,m} = \sqrt{2\pi(l^2 + m^2)} / \kappa B$$

$$\text{for } \kappa(H_0 - H_{c1}) > 0.0394. \quad (45)$$

We note that  $(\partial B/\partial H_0)_T$  is always positive, which indicates that the situation discussed is stable.<sup>7</sup>

For very large values of  $\kappa$  Eq. (35) can be used to find  $H_{c1}$ . To do this one must solve (26) and (27) numerically, after which  $\varepsilon$  is calculated from (31). On the other hand, finding  $B(H_0)$  in this case requires solving a separate and very complex problem.

To conclude this section, let us turn our attention to the intermediate state in noncylindrical specimens (or cylinders in nonparallel fields). It is most interesting that no intermediate state arises in specimens for which  $\kappa \gg 1$ . One obtains instead a uniform pattern of a mixed state (in ellipsoidal samples). When the transition in the field  $H_{c1}$  is of the first order, however, an intermediate state does arise.

### 3. COMPARISON OF THEORY AND EXPERIMENT

No detailed data exist currently on the magnetic properties of superconducting alloys. The only work which is more or less suitable for comparison with the theory is that of Shubnikov, Khotkevich, Shepelev, and Riabinin,<sup>4</sup> performed in 1937. Let us compare the theoretical results with the data of this work.

If we are to speak of general qualitative characteristics, we must first note that starting at some alloy concentration, the  $B(H_0)$  curve indicates a region of partial field penetration; this corresponds to the conclusion reached above on the existence of a mixed state. This region is bounded by the two field strengths  $H_{c1}$  and  $H_{c2}$ , and although the transition from the superconducting state to the mixed one can be either of first or of second order, the transition to the normal state must definitely be of first order. In the neighborhood of  $H_{c2}$ , the  $B(H_0)$  curve is linear. All this is in agreement with the results of Sec. 1. Further, if the alloy concentration is sufficiently high, the transition at  $H_{c1}$  seems also to be of second order. According to the

Alloy composition	T°K	$H_{c2}$	$H_{cm}$	$\kappa\sqrt{2}$	$\tan \alpha$ (theory)	$\tan \alpha$ (exp.)	$H_{c1}$ (theory)	$H_{c1}$ (exp.)
Pure Pb	4.24		540	0.22÷0.36				
Pb + 2.5% Tl	4.22	704	596	1.18	2.12	2.2	—	510
	1.92	1100	846	1.3	1.21	1.2	—	740
Pb + 5% Tl	4.22	1010	561	1.8	0.38	0.38	—	430
	1.82	1730	854	2	0.27	0.27	—	600
Pb + 15% Tl	4.22	2170	534	4.07	0.054	0.052	150	230
	1.76	3760	762	4.9	0.036	0.036	210	310
Pb + 30% Tl	4.22	2840	575	4.94	0.036	0.033	150	170
	1.70	4460	756	5.9	0.025	0.023	190	240
Pb + 50% Tl	4.22	2270	370	6.14	0.023	0.02	90	110
	2.3	4270	520	8.2	0.013	0.014	120	150
Pb + 2% In	4.22	983	604	1.6	0.51	0.45	—	530
	1.95	1520	842	1.8	0.37	0.3	—	730
Pb + 8% In	4.22	2400	563	4.26	0.049	0.045	170	220
	1.75	3780	812	4.7	0.041	0.043	240	310

theory, at this point the  $B(H_0)$  curve has a vertical tangent, and then begins gradually to approach the straight line  $B = H_0$ . This conclusion also more or less agrees with the experimental curves.

Going on to a quantitative comparison, we note that the two parameters  $H_{cm}$  and  $\kappa$  which enter into the theory must be determined from the experimental curves. This can be done by finding the upper critical field  $H_{c2}$  and the area under the curve of the magnetic moment  $B(H_0) - H_0$ , where  $H_0$  is the field strength. In fact from the definition of  $H_{cm}$ , we have

$$\frac{1}{2} H_{cm}^2 = \int_0^{\infty} [H_0 - B(H_0)] dH_0$$

$$= \int_0^{H_{c2}} [H_0 - B(H_0)] dH_0. \quad (46)$$

Of course in order that  $H_{cm}$  defined in this way have meaning, it is necessary that the experimental  $B(H_0)$  curve be that for equilibrium. When we know  $H_{cm}$ , we can find  $\kappa$  from the equation  $H_{c2} = \kappa\sqrt{2}H_{cm}$ .

The values of  $H_{cm}$  and  $\kappa$  obtained in this way can be used to compare the magnetic moment in the neighborhood of the upper critical field  $H_{c2}$  with experiment. According to Sec. 1, the dependence of  $M$  on  $H_0$  is given by Eq. (23).

We can compare with experiment, in addition to  $M$ , Eq. (36) for the first critical field.

The results of such comparison are given in the

Table.\* One should bear in mind that the accuracy with which the slope of the magnetic moment curve is determined in the neighborhood of  $H_{c2}$  (let us denote the ratio in Eq. (23) by  $\tan \alpha$ ) and  $H_{c1}$  from the curves of Shubnikov, *et al.*<sup>4</sup> is no greater than 15%. It is seen from the table that the data for  $\tan \alpha$  are in very good agreement with the theory. The values of  $\tan \alpha$  vary from specimen to specimen by a factor as high as 150, and the theory always gives the correct value within the above-mentioned limits.

The situation with respect to  $H_{c1}$  is somewhat worse. Although the theoretical values are always of the same order of magnitude as the experimental ones, they lie too low in all cases. It should be borne in mind, however, that the values of  $\kappa$  dealt with here are not very large compared to unity, and this can lead to a systematic error in determining  $H_{c1}$  from (35), an equation which is valid only when  $\kappa \gg 1$ . It is not difficult to see in which direction the theoretical value must be corrected in order to account for the fact that  $\kappa$  is not very large. This can be done by making the natural assumption that  $H_{c1} = H_{cm}$  when  $\kappa = 1/\sqrt{2}$ . Instead of this, insertion of  $\kappa = 1/\sqrt{2}$  into Eq. (36) leads to a negative value of  $H_{c1}$ . Thus for values of  $\kappa$  which are not

\* It is significant here that the theory remains applicable for temperatures far from  $T_c$ . As fields close to  $H_{c2}$ , this conclusion is strictly correct, since  $\Psi$  is small in this region. Recently, however, Ginzburg<sup>5</sup> has presented arguments which tend to show that the theory is applicable also for arbitrary fields and temperatures.

very large, the actual theoretical value of  $H_{c1}$  should be greater than that given by (36). In principle, it is also possible to perform a comparison between experiment and Eq. (45) for  $B(H_0)$  in the neighborhood of  $H_{c1}$ . This equation, however, is in principle valid only for  $\kappa \gg 1$ , so that only a qualitative comparison is possible; this has already been done at the beginning of this section. Furthermore, the experimental curves in this region are measured with very low accuracy, so that a quantitative comparison is not really very meaningful. We note here that according to the experimental data, when  $\kappa$  is close to  $1/\sqrt{2}$  the transition to the mixed state would seem to be a first order phase transition. In this case the concept of isolated filaments does not correspond at all to the mixed state at the transition point.

In order to investigate the dependence of  $H_{cm}$  and  $\kappa$  on the concentration, let us compare their values at a fixed temperature of  $4.2^\circ$ . For comparison, the table gives the data also for pure lead at the same temperature, taken from Lock.<sup>9</sup> It is seen from the table that  $H_{cm}$  hardly depends on the concentration (except for the 50% Pb-Tl alloy). The variation is no greater than 11% and has no apparent regularity. However,  $\kappa$  increases regularly with increasing concentration, and varies for Pb-Tl alloys from 1.18 at 2.5% Tl to 6.14 at 50% Tl.

Some years ago Pippard<sup>10</sup> made the interesting suggestion that there exists in superconductors a certain characteristic length (the coherence length) which determines the "range of order" of the superconducting electrons. This length characterizes the minimum distance at which the number of superconducting electrons may change significantly. It may, for instance, refer to the thickness of the transition layer between the normal and superconducting phases.

According to the theory of the previous sections, the quantity  $\delta/\kappa$  should play the role of such a length. This length determines the thickness of the transition layer in the intermediate state.<sup>2</sup> If we bear in mind that  $\kappa = (\sqrt{2}e/hc)H_{cm}\delta^2$ , then it is clear that the characteristic length is proportional to  $1/\sqrt{\kappa}$ . According to Pippard, this length should depend strongly on the impurity concentration. At high concentrations it should be of the order of the mean free path of an electron in the metal. It is seen from the table that this concept is not in disagreement with the experimental data.

The results described show that the quasi-microscopic theory is able to describe many of the regular properties of superconducting alloys. Thus these regular properties arise primarily not because macroscopic regions with relatively large values of  $T_c$  and  $H_{cm}$  arise in the alloy, as has been thought previously, but from the destruction of electron order at distances small compared with the penetration thickness  $\delta$ .

It is true, of course, that macroscopic inhomogeneities may be of importance. Without even going into the fact that a strong inhomogeneity can change the overall pattern, we note that such a phenomenon as the remanent magnetic moment cannot be described by the present theory and is more probably a result of such inhomogeneities. The structure of the field penetration itself should be extremely sensitive to inhomogeneities, so that in real substances it would hardly be in the form of a regular lattice.

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