Angular Dependence of the Superconductive Nucleation Field H_{c4}

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The angular dependence of the superconducting nucleation field of a system with two intersecting vacuum faces is investigated. Within the framework of the simple Ginzburg-Landau theory using a trial-function approach, it is shown that superconductivity can nucleate along the edge of a wedge-shaped geometry in fields H_{c4} above H_{c3}. This nucleation field is calculated for different angles of the wedge and different directions of the field. The effect is studied experimentally on evaporated films with wedge-shaped edges produced with the use of a shadowing technique. It is shown that the experimental results can be explained on the basis of a H_{c4} model.

I. INTRODUCTION

The surface superconductivity in fields greater than H_{c3} has recently attracted a great deal of interest.[1] Since the discovery of Saint-James and de Gennes[2] that the presence of a vacuum interface enhances the nucleation field, it was obvious to suspect that two intersecting vacuum interfaces might enhance the nucleation field even more. Houghton and McLean[3] and van Gelder[4] have calculated this nucleation field for a wedge-shaped geometry, which was referred[4] to as H_{c4}. They find that for small values of the angle 2α between the vacuum interfaces

$$H_{c4} \geqslant (\tfrac{1}{2}\sqrt{3}\,\alpha) H_{c2} , \qquad (1)$$

where H_{c2} is the bulk-nucleation field $\sqrt{2}\kappa H_c$ [κ is the Ginzburg-Landau (GL) parameter, H_c the thermodynamic critical field]. It has been emphasized by Fink[5] that H_{c4} has nothing to do with a new mechanism of nucleation; H_{c4} lies entirely within the framework of the GL theory,[6] which for an infinite slab of thickness d, with the applied field parallel to the surface planes in the limit $d \ll \xi$, where ξ is the GL coherence length, gives a nucleation field

$$H_n = (12)^{1/2} (\xi/d) H_{c2} . \qquad (2)$$

As has been noted by Fink,[5] a wedge-shaped specimen with almost parallel surfaces can be approximated by a slab with an effective thickness $\bar{d}$ and then Eqs. (1) and (2) are equivalent. A variational calculation for $\bar{d}$ as a function of α was carried out by van Gelder.[4] It is intuitively clear and follows from the calculations that superconductivity nucleates along the wedge in a "line" of thickness of the order ξ. It is the purpose of the present paper to study the dependence of H_{c4} as a function of the angle between the wedge and the applied field, and to look into the possibility of observing H_{c4} experimentally.[7]

II. CALCULATION OF ANGULAR DEPENDENCE OF H_{c4}

Our calculations are based on the variational formulation[8] of the problem of nucleation of superconductivity. We consider the Ginzburg-Landau[6] Gibbs free energy ΔG between the superconducting and the normal phases. Nucleation becomes possible when ΔG vanishes, i.e.,

$$\kappa^{-2} \int d\vec{r}\, (\vec{\nabla} f)^2 + \int d\vec{r}\, [(\vec{A} + \kappa^{-1} \vec{\nabla}\Phi)^2 - 1] f^2 = 0 . \qquad (3)$$

Here, $f(\vec{r})$ is the modulus of the order parameter and $\Phi(\vec{r})$ is its phase. As a unit of length, we use the weak-field penetration depth, such that the magnetic field varies typically like $e^{-\vec{r}\cdot\vec{n}}$ near a vacuum interface while $\vec{n}$ is the unit vector normal to the surface. The vector potential $\vec{A}(\vec{r})$ is chosen to represent a homogeneous applied static magnetic field $\vec{H} = \mathrm{rot}\vec{A}$ of arbitrary direction. This choice is the same as in Ref. 8 and is only motivated for a study of the *onset* of nucleation. The unit for $\vec{A}$ and hence for $\vec{H}$ is such that the bulk critical field $H_c = 1/\sqrt{2}$. The system which we consider is bounded by the planes characterized by $\varphi = \pm\alpha$ in cylindrical coordinates $\{r, \varphi, z\}$ (Fig. 1). Along the x, y and z directions, the applied field has the following components:

$$H_x = H \sin\gamma \cos\beta ,$$
$$H_y = H \sin\gamma \sin\beta , \qquad (4)$$
$$H_z = H \cos\gamma .$$

The angle between $\vec{H}$ and the z axis is γ, and the angle with the xz plane ($\varphi = 0$) is β. We choose a gauge where this field is represented by the vector potential $\vec{A}$ given by

$$A_r = 0 ,$$
$$A_\varphi = \tfrac{1}{2} H r \cos\gamma , \qquad (5)$$
$$A_z = H r \sin\gamma \sin(\varphi - \beta) .$$

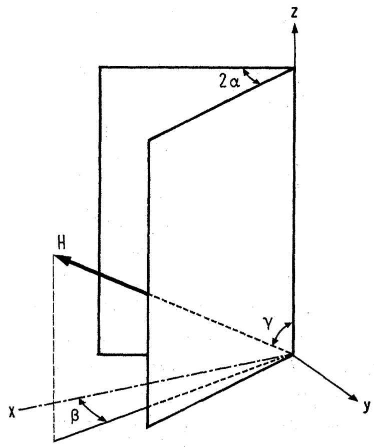

FIG. 1. Geometrical situation.

A reduction of the unit of length by a factor $(\kappa H)^{1/2}$ in Eq. (3) gives

$$\int d\vec{r}\,(\vec{\nabla} f)^2+\int d\vec{r}\left[\left(\frac{\partial\Phi}{\partial r}\right)^2+\left(\tfrac{1}{2}r\cos\gamma+\frac{1}{r}\frac{\partial\Phi}{\partial\varphi}\right)^2+\left(r\sin\gamma\sin(\varphi-\beta)+\frac{\partial\Phi}{\partial z}\right)^2-E\right]f^2=0\;, \qquad (6)$$

where E is defined as

$$E=\kappa/H=H_{c2}/H. \qquad (7)$$

Since the current density components normal to the vacuum interfaces must vanish, we get for $f(\vec{r})$ and $\Phi(\vec{r})$ the boundary conditions

$$\left.\frac{\partial f}{\partial\varphi}\right|_{\varphi=\pm\alpha}=\left.\frac{\partial f}{\partial r}\right|_{r=0}=\left.\frac{\partial\Phi}{\partial r}\right|_{r=0}=0$$

and (8)

$$\left.\frac{\partial\Phi}{\partial\varphi}\right|_{\varphi=\pm\alpha}=-\tfrac{1}{2}r^2\cos\gamma\;.$$

The variational method gives an upper bound for the smallest value of E for which Eq. (3) is satisfied and nucleation along the wedge can occur. For this purpose we substitute the following trial functions into Eq. (6):

$$f(\vec{r})=f(r),\quad \Phi(\vec{r})=-\tfrac{1}{2}r^2\alpha F(\varphi/\alpha)+mz\;, \qquad (9)$$

with m a constant and, in order to satisfy the boundary conditions, $F'(\pm 1)=\cos\gamma$. This choice of trial functions is intended to study the possibility of nucleation along a wedge *only*, and is not expected to describe nucleation at the plane vacuum interfaces $\varphi=\pm\alpha$ at H_{c3}. The ansatz of Eq. (9) is only useful for small values of α; in fact, if $2\alpha \gtrsim 76°$,[4] the nucleation fields are not expected to exceed H_{c3} so that this nucleation can only occur at the surfaces $\varphi=\pm\alpha$. Substitution of Eq. (9) into Eq. (6) gives

$$\int_0^\infty dr\left[\left(\frac{df}{dr}\right)^2 r+Pf^2r^3-2mQf^2r^2+(m^2-E)f^2r\right]=0\;, \qquad (10)$$

with

$$P=\tfrac{1}{2}\alpha^2\int_{-1}^{+1}F^2(x)\,dx+\tfrac{1}{8}\int_{-1}^{+1}[F'(x)-\cos\gamma]^2dx+\tfrac{1}{2}\sin^2\gamma\{1-[(\sin 2\alpha)/2\alpha]\cos 2\beta\}\;, \qquad (11)$$

$$Q=\sin\gamma\sin\beta\sin\alpha/\alpha\;. \qquad (12)$$

If $\gamma=0$, as in the case of Refs. 3 and 4, it is possible to show analytically, that the lowest value of E for which Eq. (10) is satisfied corresponds to a function $f(r)$ which is of the form $\exp[-\frac{1}{2}(r/1a)^2]$, where $1a$ is a constant. The problem of finding the smallest value of E for which Eq. (10) is satisfied is equivalent to minimizing the functional on the left-hand side of Eq. (10) with respect to f, considering E as a Lagrange multiplier. If $\gamma=0$, E attains a minimum when $m=0$. Variation of Eq. (10) with respect to f leads to the condition that

$$u\frac{d^2g}{du^2}+(1-2u)\frac{dg}{du}+2Mg=0\;, \qquad (13)$$

where

$$u=\tfrac{1}{2}r^2\sqrt{P}\;, \qquad (14)$$

$$g(u)=e^u f(r)\;, \qquad (15)$$

$$M=-\tfrac{1}{2}+E/4\sqrt{P}\;. \qquad (16)$$

Equation (13) has two solutions, one of which is singular at $u=0$, because the Wronskian is e^{2u}/u. The other solution increases exponentially like e^{2u} as $u\to\infty$, unless M is a positive integer or zero. The lowest value of E corresponds to the eigenvalue $M=0$ for which $g(u)=1$. Larger eigenvalues [e.g., $M=1$, $g(u)=1-2u$] suggest the existence of metastable normal solutions below H_{c4}, nucleation becoming possible at $H=H_{c4}/(2M+1)$ where $M=0, 1, 2, \cdots$.

It is therefore reasonable to choose for $f(r)$ a function of the type $\exp[-\frac{1}{2}(r/a)^2]$, whether γ is zero or not. This gives

$$E=1/a^2+Pa^2-mQa\sqrt{\pi}+m^2. \qquad (17)$$

Therefore, the smallest value of E is

$$E=(4P-\pi Q^2)^{1/2}\;. \qquad (18)$$

Q can be calculated for a given orientation of $\vec{H}$, but P is still a functional of F, and E has to be minimized with respect to this functional. Making use of the boundary conditions for F, we get

$$F(x)=\frac{\cos\gamma}{2\alpha}\,\frac{\sinh 2\alpha x}{\cosh 2\alpha}\;. \qquad (19)$$

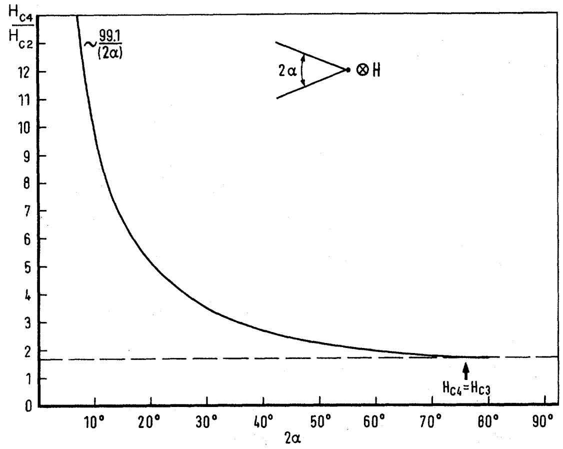

FIG. 2. Calculated H_{c4}/H_{c2} as a function of the wedge angle for $\gamma = 0$.

Consequently P becomes

$$P = \tfrac{1}{4}\cos^2\gamma\left(1 - \frac{\tanh 2\alpha}{2\alpha}\right) + \tfrac{1}{2}\sin^2\gamma\left(1 - \frac{\sin 2\alpha}{2\alpha}\cos 2\beta\right) \quad . \tag{20}$$

A lower bound for the nucleation field H_{c4} is hence found to be

$$(H_{c4}/H_{c2}) = E^{-1} = (4P - \pi Q^2)^{-1/2} , \tag{21}$$

where P and Q are functions of α, β, and γ [Eqs. (20) and (12)]. In Figs. 2–4, H_{c4}/H_{c2} is plotted for several geometrical situations. It is interesting to note that for $\beta = 0°$ and for small angles of α, H_{c4} depends very little on γ.

III. EXPERIMENTAL RESULTS

It is clear from our trial-function solution that superconductivity just below H_{c4} should be localized along the wedge, roughly within the GL coherence length around the edge. Therefore, the radius of curvature of the edge ("sharpness" of the edge) should be smaller than, say, ~1000 Å. Preliminary investigations on mechanically cut bulk superconductors failed to show any reproducible effects.[9] We decided therefore to look into the edges of thin films.

The films are prepared in the form of strips by vacuum evaporation of indium at a pressure of about 10^{-6} Torr onto microscope slides through a mask which was slightly removed from the substrate. Because of the well-known shadowing effect,[10] the edges of the film are not completely sharp but show a wedgelike geometry of the type we would like to study (Fig. 5). The thickness of the films was measured interferometrically, using a Varian Å-scope multiple-beam interferometer.

As a first step, we repeated the well-known experiment[10] of measuring the parallel ($\gamma = 90°$, $\beta \simeq 0°$) and perpendicular ($\gamma = 90°$, $\beta \simeq 90°$) critical field of such a wedge-shaped film. In order to compare with the critical fields of the film without a wedge, two identical films were evaporated simultaneously next to each other, and one of them was trimmed with a razor blade. In Figs. 6 and 7, we show recorder tracings of the resistance transition vs magnetic field in the parallel and perpendicular positions for a trimmed and an untrimmed film. There is an obvious difference in the critical fields of the two films in the case with the field parallel to the films, while in the perpendicular case the two critical fields seem to be equal. These well-known results can be interpreted with the theory of Sec. II according to which the film edges are superconducting until H_{c4} is reached. Ideally, the edges of the trimmed film have two

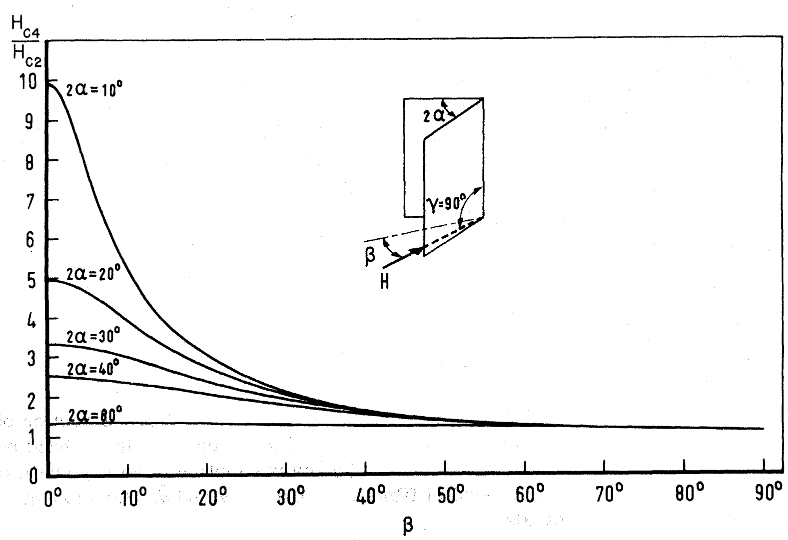

FIG. 3. Calculated angular dependence of H_{c4}/H_{c2} for $\gamma = 90°$.

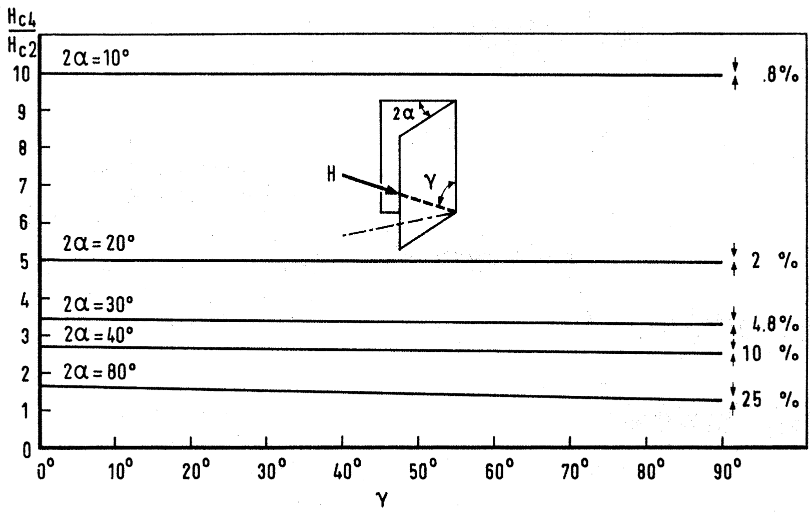

FIG. 4. Calculated angular dependence of H_{c4}/H_{c2} for $\beta = 0$.

wedges for which $2\alpha = 90°$. The field directions for the parallel and perpendicular cases correspond to $\gamma = 90°$, $\beta = 45°$ for these wedges. However, as can be seen from Fig. 3, the critical field of these wedges does not exceed the critical field H_{cf} of the film. The latter field, as given by the Tinkham theory[11, 12] for thin films (although one might argue about the applicability of this theory for our rather thick films), is $H_{cf\perp} = H_{c2}$ for the perpendicular case, whereas it is given by $H_{cf\parallel} = H_n$ [Eq. (2)] for the parallel one. The situation is quite different for the untrimmed film (α small) in a parallel field ($\beta \sim 0°$), because here the nucleation field H_{c4} of Eq. (21) exceeds that of the film. This prediction seems to be confirmed by the experiment (Fig. 7). For the parallel position in the trimmed case, the critical field $H_{cf} = H_n$ is given by Eq. (2), while in the untrimmed case, according to Eq. (21), $H_{c4}/H_{c2} = E^{-1}(\alpha, \beta = 0°, \gamma = 90°)$. Using the measured values of H_{c4} and H_{c2} and Fig. 3, we get an angle of $2\alpha \simeq 30°$. An attempt to measure the angle 2α interferometrically was not very successful, owing to a fuzzing out of the film at the edge [Fig. 5(c)]. $2\alpha'$ seems to be of the order of $1°$, while 2α might be of the order of $10°$. We realize of course that this does not in any way prove the existence of something like H_{c4}; everything could equally well be explained by assuming that the edge corresponds to a steplike geometry as shown in Fig. 5(b) and as suggested by Fink.[5] This would correspond to two (or more) parallel films with thicknesses $\bar{d}$ and d, where the critical fields are given by Tinkham-type[11, 12] formulas. However, this would no longer be true if we vary the angle γ: For a film, there should be no difference in the critical field if the field moves in the plane of the film; for the wedge geometry however, Fig. 4 shows that there is a variation of H_{c4} with γ.

In order to check this possibility, we evaporated a square film as sketched in Fig. 8, again using the shadowing technique by slightly removing the mask. The current I was passed through one diagonal of the square, while the voltage V was measured over the other diagonal. Owing to the symmetry of the configuration, there should be no voltage if the square is entirely normal or entirely superconducting. However, if part of the film is normal, the symmetry is distorted, the corners are no longer equipotential points, and a voltage should be detectable. If again we assume our H_{c4} model to be valid for this square film (i.e., homogeneous with wedge-shaped edges), for the edges a and b, we have $\gamma = 90°$ and β is variable, while for the edges c and d, γ is variable and $\beta \simeq 90°$. The value of β for a and b is equal to the value of γ for c and d (Fig. 8). Using our trial-function solutions, we

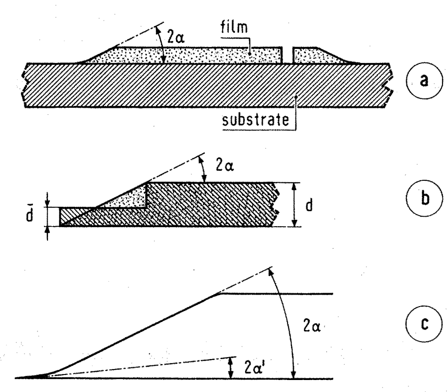

FIG. 5. Edges of films. (a) Typical cross section of evaporated film, showing tapered edges. Right-hand edge has been trimmed. (b) Approximation of an edge with two plane parallel films of thickness d and $\bar{d}$. (c) Fuzzing out of edge.

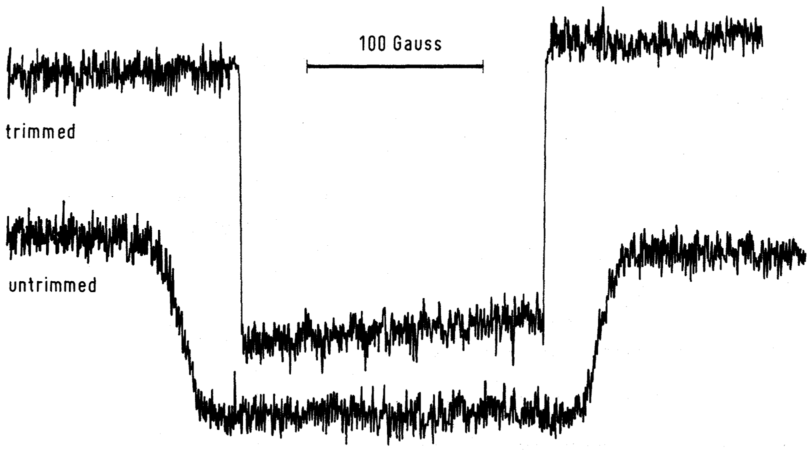

FIG. 6. Recorder tracings of resistance transition vs magnetic field for a trimmed ($\beta \simeq 45°$) and untrimmed ($\beta \simeq 0°$) indium film with the field in the plane of the film ($\gamma = 90°$). Measuring current $I = 5\ \mu A$, temperature $T = 0.84 T_c$, thickness $d \simeq 7500$ Å.

can calculate H_{c4}/H_{c2} as a function of α and β. This is shown in Fig. 9; as can be seen from this figure, H_{c4} (cd) is bigger than H_{c4} (ab). If β increases, the difference between the two values for H_{c4} gets smaller. For values of $\alpha \simeq 10°$ (which should roughly correspond to the experimental situation) at angles of $\beta \simeq 60°$, the difference is smaller than about 0.5% and should no longer be detectable with our experimental setup. In Fig. 10, we show recorder tracings of the voltage over the diagonal versus the magnetic field for different values of angle β. There is definitely an asymmetry which leads to peaks in the signal; the critical fields decrease as β increases, and for angles of the order of $\beta \gtrsim 60°$, no asymmetry in the square film can be detected any more.

We would like to emphasize once again that this is certainly no conclusive proof of the existence of H_{c4}. Small inhomogeneous parts in the inside of the film would also lead to asymmetric situations which result in signals. By trimming the edges of the square film, we could reduce the signals; however, we were unable to create an entirely symmetric situation (i.e., no signals for all fields and all values of β) for trimmed films.

In order to check the angular dependence of H_{c4} quantitatively and to discriminate against the angular dependence of the critical field of the "bulk" film, we measured the two critical fields (of the film or the edge) simultaneously. The critical field of the interior of the film was measured using a tunneling-junction geometry (Fig. 11). First we evaporated an aluminum film as one side of the junction and let it oxidize. On top of it we evaporated the indium film to be studied, again using the shadowing technique. The Al-Al_2O_3-In junction has then the whole interior of the film to be studied as one electrode; this junction should then measure an averaged bulk property of this film. The critical field of the film was determined by measuring the derivative (dV_T/dI_T) at zero bias $V_T = 0$ as a function of the magnetic field. At the

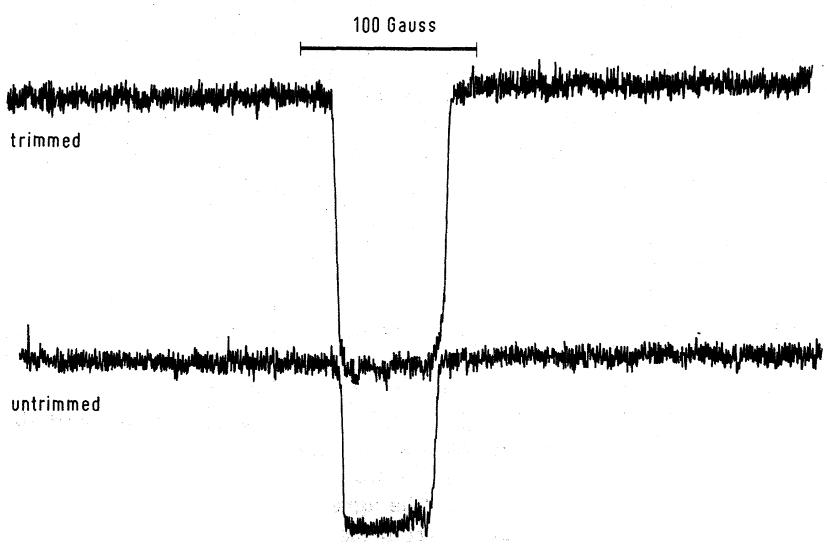

FIG. 7. Recorder tracings of resistance transition vs magnetic field for a trimmed ($\beta \simeq 45°$) and an untrimmed ($\beta \simeq 90°$) indium film with the field perpendicular to the plane of the film ($\gamma = 90°$). Measuring current $I = 5\ \mu A$, temperature $T = 0.84 T_c$, thickness $d \simeq 7500$ Å.

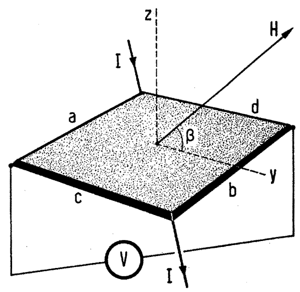

FIG. 8. Geometrical situation for a square film.

same time, the critical field of the edge was measured with the simple dc technique by passing a current I_R and measuring the voltage V_R. All measurements were done at temperatures where the aluminum film was in the normal state. During the measurements, the magnetic field was always perpendicular to the edges of the film ($\gamma = 90°$), while the angle β was varied between 0° (position of the field parallel to the film) and 90° (position of the field perpendicular to the film). For the simple dc resistance measurements, we

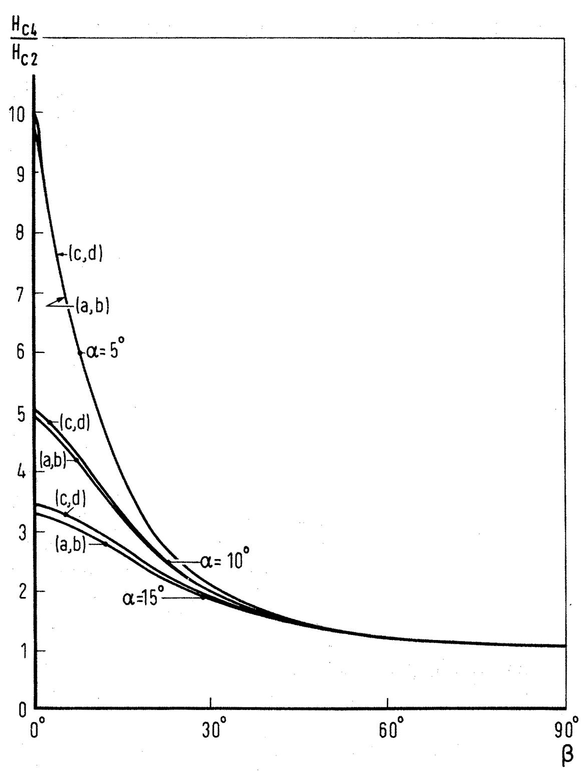

FIG. 9. Theoretical curves of H_{c4} as a function of β for the two pairs of edges (a b) and (c d) of Fig. 8 for different values of the wedge angle α.

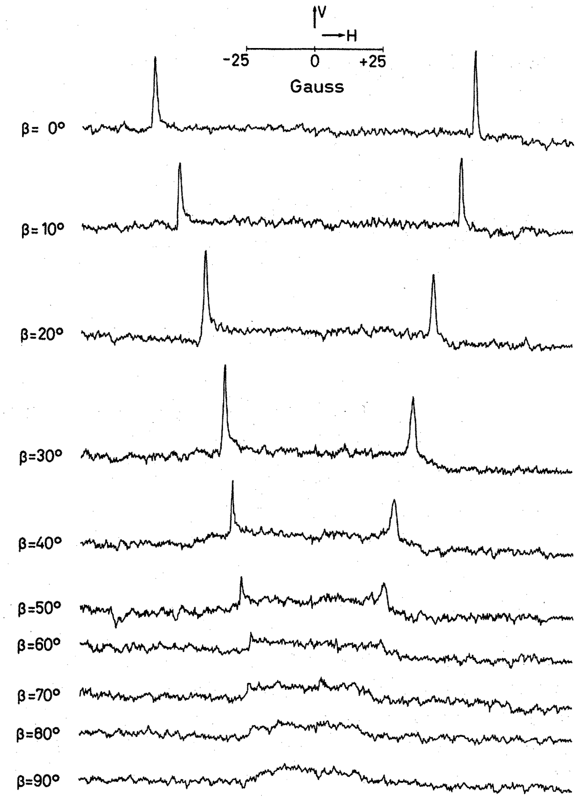

FIG. 10. Recorder tracings of the diagonal voltage of a square indium film as a function of the magnetic field for different angles β. Measuring current $I = 0.5$ mA, temperature $T = 0.86 T_c$, thickness $d \simeq 8000$ Å.

expect an angular dependence of the critical field of the edges as given by our H_{c4} model [Eq. (21), Fig. 2]:

$$H_{c4}/H_{c2} = [E(2\alpha, \beta, \gamma = 90°)]^{-1} , \qquad (22)$$

while for the tunneling measurements, the angular dependence of the critical field $H_{cf}(\beta)$ of this film should be given by a Tinkham-type[11, 12] formula:

$$\frac{H_{cf}(\beta)\sin\beta}{H_{cf\perp}} + \left(\frac{H_{cf}(\beta)\cos\beta}{H_{cf\parallel}}\right)^2 = 1 , \qquad (23)$$

where $H_{cf\perp}$ is the perpendicular and $H_{cf\parallel}$ the parallel critical field of the film.

In Fig. 12, we plotted the critical fields as measured with tunneling as a function of the angle β. The theoretical curve H_{cf} is given by Eq. (23) and adjusted at the two points $H_{cf\parallel} = H_{cf}(0°)$ and $H_{cf\perp} = H_{cf}(90°)$. The agreement is satisfactory, as is well known from the extensive studies of Harper

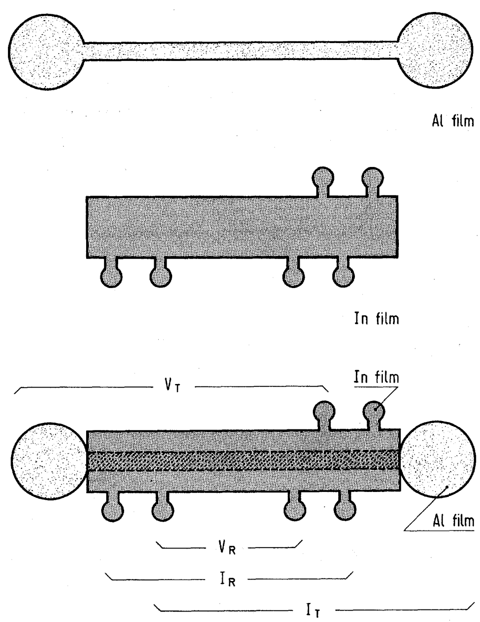

FIG. 11. Tunneling setup to measure the critical field of the edge H_{c4} and the critical field of the film H_{cf} simultaneously. Tunneling: voltage over V_T, current through I_T; dc resistance measurements: voltage over V_R, current through I_R.

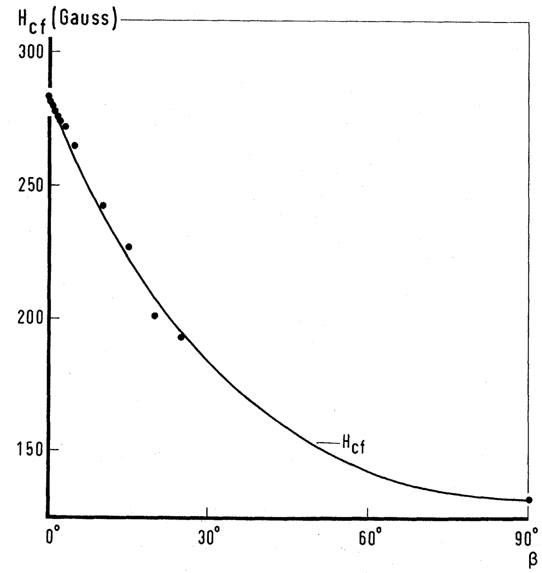

FIG. 12. Experimental values of the critical field of the film as measured with the tunneling junction. The curve labeled H_{cf} is calculated from Eq. (23) and adjusted for $\beta = 0°$ and $\beta = 90°$.

and Tinkham.[12] As another check, the thickness d of the film should be given by[11]

$$d = \left(\frac{6\varphi_0}{\pi} \frac{H_{cf\perp}}{H_{cf\parallel}^2}\right)^{1/2}, \tag{24}$$

where φ_0 is the quantum of flux. Using our experimental values of $H_{cf\perp}$ and $H_{cf\parallel}$, we get $d \simeq 2500$ Å, while the interferometric measurements lead to a value of $d \simeq 2800$ Å. Again, the agreement is satisfactory.

In Fig. 13, we plot the critical fields as determined by the simple dc resistance measurements. In the same figure, we also plot a theoretical $H_{c4}(\beta)$ curve, adjusted at the two points $H_{c4}(0°)$ and $H_{c4}(90°)$. The agreement is reasonable, and the resulting angle of $\alpha \simeq 16.7°$ is not unrealistic (an attempt to measure α interferometrically again failed due to the fuzzing out at the edge). The theoretical curve marked H_{cf} is a Tinkham-type relation [Eq. (23)], again adjusted at the two points $H_c(0°)$ and $H_c(90°)$. The agreement is somewhat less satisfactory, which means that our results can better be explained by assuming a wedge-shaped geometry instead of a simple film geometry. It has to be noted that an absolute measurement of the critical field of a film using our tunneling technique is very inaccurate, because the superconducting normal transition curve is very broad owing to the gapless situation; this is not very serious for relative measurements such as angular dependence, where the same well-defined (say 50% of the transition), but somehow arbitrary point on the transition curve can always be used to define an $H_c(\beta)$. However, this does not make a comparison between

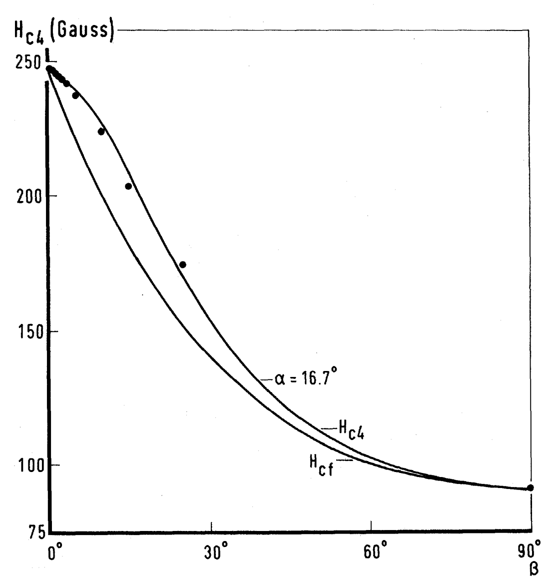

FIG. 13. Experimental values of the critical field of the edge of a film as measured with dc. The curve labeled H_{c4} is calculated from Eq. (21), and the curve H_{cf} is calculated from Eq. (23); both are adjusted at $\beta = 0°$ and $\beta = 90°$.

measured absolute values of H_{ct} and H_{c4} very meaningful.

In conclusion, we would like to note that we certainly do not believe we have proved experimentally the existence of something like H_{c4} unambiguously. Most of our experiments can be explained by assuming inhomogeneous parts in the bulk of the films. However we think we have shown that wedge-shaped geometries behave differently from film geometries in magnetic fields, and it may even be that inhomogeneous parts can be considered as wedge shaped, which would correspond to an H_{c4} model.

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[1]For a recent review see, e.g., W. C. H. Joiner, Phil. Mag. 20, 807 (1969).

[2]D. Saint-James and P. G. de Gennes, Phys. Letters 7, 306 (1963).

[3]A. Houghton and F. B. McLean, Phys. Letters 19, 172 (1965).

[4]A. P. van Gelder, Phys. Rev. Letters 20, 1435 (1968).

[5]H. J. Fink, Phys. Rev. 177, 1017 (1969).

[6]V. L. Ginzburg and L. D. Landau, Zh. Eksperim. i Teor. Fiz. 20, 1064 (1950).

[7]For a preliminary report on these investigations see A. P. van Gelder, J. W. Hendriks, and P. Wyder, in *Proceedings of the Eleventh International Conference on Low Temperature Physics, St. Andrews*, 1968, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrew's U. P., St. Andrew's, Scotland, 1969), Vol. II, p. 956.

[8]P. G. de Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966).

[9]H. N. de Lang, Doctoraalscriptie, University of Nijmegen, The Netherlands, 1969 (unpublished).

[10]See, e.g., V. L. Newhouse, *Applied Superconductivity* (Wiley, New York, 1964).

[11]M. Tinkham, Phys. Rev. 129, 2413 (1963).

[12]F. E. Harper and M. Tinkham, Phys. Rev. 172, 441 (1968).

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Neutron Scattering Study of the Lattice-Dynamical Phase Transition in Nb_3Sn†

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Neutron scattering experiments have been carried out on a single crystal of Nb_3Sn through the lattice-dynamical phase transition at $T_m = 45\,°K$. A small tetragonal lattice distortion, $a/c = 1.0062$ at 4 °K, was previously established by x-ray studies, but sublattice displacements below T_m have remained undetermined. The present study reveals that the tetragonal phase exhibits *new* Bragg reflections, which are forbidden by symmetry in the cubic phase. From the intensity distribution among these new reflections, the structure was determined uniquely as D_{4h}^9 with Nb displacements from the special positions of 0.016(3) Å at 4 °K. Only the Nb sublattices shift, and in a pattern identical with the eigenvectors of the $\Gamma_{12}(+)$ $q = 0$ optic-phonon mode in the cubic phase. Such a mode is *linearly* coupled with the soft [110] shear acoustic mode. This linear coupling requires, and our measurements confirm, that the intensities of new Bragg peaks are proportional to $(a/c - 1)^2$. An optic-phonon instability is *not* required to explain these internal displacements.

I. INTRODUCTION

In recent years, extensive investigation has been carried out on many lattice-dynamical phase transitions. One of the most fascinating phase transitions known of this type occurs in high-temperature superconductors with the β-W structure (type A-15).[1] This phase transition takes place, on cooling, before the onset of the superconducting state. It is accompanied by a remarkable elastic softening, in particular, for shear modes with wave vector $\vec{q} \parallel [110]$ and polarization vector $\vec{e} \parallel [1\bar{1}0]$. For example, in the case of Nb_3Sn, the acoustic velocity[2] falls from a normal room-temperature value to near zero around $T_m = 45\,°K$. The crystal is cubic above T_m and becomes tetragonal[3] below T_m with $a/c = 1.0062$ at 4 °K.

Many experimental and theoretical studies have been published[4] on this phase transition in Nb_3Sn, as well as on the similar transition in V_3Si at 21 °K. There are, nevertheless, fundamental questions concerning the nature of the transformation which remain unsolved. Anderson and Blount[5] pointed out that if the phase change is truly of second order the tetragonal strain cannot be the primary-order parameter, and they raised the pos-