alloy systems. A more comprehensive study is currently in progress.

<sup>1</sup>P. Lederer and D. L. Mills, Phys. Rev. <u>165</u>, 837 (1968); S. Engelsberg, W. F. Brinkman, and S. Doniach, Phys. Rev. Letters 20, 1040 (1968).

<sup>2</sup>A. I. Schindler and C. A. Mackliet, Phys. Rev. Letters <u>20</u>, 15 (1968); G. Chouteau, R. Fourneau, R. Fourneaux, K. Gobrecht, and R. Tournier, Phys. Rev. Letters <u>20</u>, 193 (1968). <sup>3</sup>E. Bucher, W. F. Brinkman, J. P. Maita, and H. J. Williams, Phys. Rev. Letters <u>18</u>, 1125 (1967).

 ${}^{4}$ F. R. de Boer, C. J. Schinkel, J. Biesterbos, and S. Proost, in Proceedings of the Conference on Magnetism and Magnetic Materials, New York, 18-21 November 1968 (to be published).

<sup>5</sup>G. G. Low and T. M. Holden, Proc. Phys. Soc. (London) <u>89</u>, 119 (1966), and references therein; T. J. Hicks, T. M. Holden, and G. G. Low, J. Phys. C:

Phys. Soc. (London) Proc. 1, 528 (1968).

<sup>6</sup>B. Mozer, D. T. Keating, and S. C. Moss, Phys. Rev. <u>175</u>, 868 (1968).

<sup>7</sup>S. A. Ahern, M. J. C. Martin, and W. Sucksmith, Proc. Roy. Soc. (London), Ser. A. 248, 145 (1958).

## MAGNETOSTRICTION DUE TO SURFACE CURRENTS IN TYPE-II SUPERCONDUCTORS

G. Brändli and R. Griessen

Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule, Zürich, Switzerland (Received 20 January 1969)

The mixed state behavior of type-II superconductors is determined by both the fluxline density and the superconductive surface currents. These currents give rise to a new magnetostrictive term that depends on the surface currents <u>and</u> the flux-line density. The new term is found to be very large in the indium alloys investigated. The experimental results are explained theoretically and it is shown that the elastic constants can be calculated from measurements of the new magnetostrictive term.

Large diamagnetic or paramagnetic surface currents can be induced in superconductors of the second kind when they are in the mixed state. We have found that in indium alloys these currents give rise to dimensional changes that are comparable with the total change between the normal and superconducting states and much larger than those resulting from magnetostriction in the Meissner phase. This new and unexpectedly large effect can be explained, if the sample is regarded as composed of two phases. One, the volume phase, defines the magnetic field and the field direction just inside the surface by the value of its magnetization. The second phase is the current-carrying surface layer that is exposed to field pressures from both sides. The magnetostriction arising from the surface currents is an elastic deformation under these pressures and it yields a novel method for the determination of the elastic constants. In this note we summarize our experimental results and their theoretical explanation.

When an ellipsoidal specimen is taken into the mixed state by increasing the field H from zero or reducing it from  $H > H_{c2}$  and the change in field is then reversed, then screening currents in the surface are induced reversibly. This is

shown in the magnetization curves of Figs. 1(c) and 1(d). The short lines indicated by pairs of arrows in the figure are such reversible changes in magnetization. Their slope shows that the whole change in external field is screened from the interior of the sample by the induced currents. That the screening is really complete has



FIG. 1. Magnetostriction and magnetization curves for a flat ellipsoid made from polycrystalline In-14at.% Tl. The length change is measured at a radius of the equator. The magnetic field was applied in the equatorial plane in directions parallel (a) and (c) and perpendicular (b) and (d) to the measured radius.

<sup>\*</sup>Attached from Imperial College, London, England. †Guggenheim Fellow on leave from the General Electric Research and Development Center during 1967-1968.

recently been shown by thermodynamic arguments.<sup>1</sup>

During these magnetization changes we observed the length changes shown in Figs. 1(a) and 1(b). One notices that the length changes have opposite slopes in the two figures. A discussion of the curves is given below.

The specimen investigated was made from polycrystalline indium with 14-at.% thallium and is a flat ellipsoid of revolution with an axial ratio of 1:10. The changes of radius of the equatorial plane parallel and perpendicular to a field applied in this plane were recorded using the capacity method described by White.<sup>2</sup> The magnetization was recorded simultaneously by integration of the voltage in a pickup coil.

We have measured and discussed the magnetostriction in type-II superconductors in the absence of surface currents elsewhere<sup>3,4</sup> and we have shown that its reversible part is made up of three contributions proportional to  $\partial H_c/\partial p$ ,  $\partial \kappa/\partial r$  $\partial p$ , and  $\partial V/\partial p$  ( $H_c$  is the critical field,  $\kappa$  the Ginzburg-Landau parameter, V the volume, and p the pressure). To explain the present observations, we imagine our specimen to consist of a core of reversible material with a surface capable of carrying superconductive screening currents up to a certain critical value.<sup>5-7</sup> Since the specimen is relatively large (diameter of 30 mm in the equatorial plane), it is unnecessary to consider the current distribution in detail and we simple assume a net current just at the surface.

The starting point of our calculation is an ellipsoid in the mixed state without surface currents. The field at infinity parallel to a main axis is  $H_e$ , the demagnetization factor is D, and the permeability is  $\mu \equiv B_i/H_i$ , where  $B_i$  and  $H_i$  are the local fields inside the specimen and averaged over the flux-line structure. If the external field is now changed in the correct sense by an amount  $\Delta H_e$ , surface currents are induced so as to screen the interior of the sample completely.

The field inside the sample is then constant and parallel to  $H_e$ , and has the value  $H_i = H_e / [1-D(1 -\mu)]$ . The field just outside the surface can be calculated using the continuity conditions for the field components parallel and perpendicular to the surface. One has to notice that only the field component parallel to the surface can be screened by a surface current. For this component the outer field is greater than in the current free state by a factor  $1+\Delta H_i/H_i$ , where  $\Delta H_i = \Delta H_e / (1-D)$  is the field screened by the surface currents. The normal component is independent of  $\Delta H_e$ .

The field inside and outside the surface can therefore be calculated exactly. The Maxwell stress-tensor difference for the two fields gives the pressure acting on a surface element in the two states with and without screening currents. The pressure lies in the plane given by the inner field and the normal to the surface element and can be divided into two components:  $p_{\parallel}$  parallel to  $H_i$ , and  $p_{\perp}$  perpendicular. These are given by

$$\Delta p_{\parallel} = -H_{i} \Delta M (1-\mu) \sin^2 \varphi \cos \varphi, \qquad (1a)$$

$$\Delta p_{\perp} = -H_{i} \Delta M (\sin^{3}\varphi + \mu \sin\varphi \cos^{2}\varphi), \qquad (1b)$$

where  $\Delta M = -\Delta H_i/4\pi$  is the magnetization resulting from the induced surface currents and  $\varphi$  is the angle between the inner field and the normal to the surface element.

The evaluation of the elastic deformations under the stress distribution so imposed is complicated. One can, however, show that the deformations always have the form

$$\Delta r_{\parallel} = r_{\parallel} H_i \Delta M [c_{\parallel} (1-\mu) - 2\sigma] / E, \qquad (2a)$$

$$\Delta r_{\perp} = r_{\perp} H_i \Delta M [-c_{\perp} (1-\mu)\sigma + 1-\sigma]/E, \qquad (2b)$$

where  $r_{\parallel}$  and  $r_{\perp}$  are the radii of the equator of the flat ellipsoid measured parallel and perpendicular to  $H_{\rho}$ , and  $\sigma$  and E are the Poisson number and Young's modulus, respectively. The difficulty lies in the evaluation of the constants  $c_{\parallel}$ and  $c_{\perp}$ , which depend only on the form of the specimen and the Poisson number of the material used. We have studied approximate solutions for a general ellipsoid of revolution and found that the constants c are of the order of unity. For an infinitely flat ellipsoid,  $c_{\parallel}$  and  $c_{\perp}$  tend to unity and, if further  $\mu = 0$  as in the Meissner state and at  $H_{c1}$ , then the brackets in formulas (2a) and (2b) tend to  $1-2\sigma$ . Therefore an infinitely flat ellipsoid (not a film!) is isotropically compressed in the Meissner state by a negative  $\Delta M$ , i.e., a positive  $\Delta H_e$ . Near  $H_{c2}$ , on the other hand,  $\mu$  is close to unity, and  $\Delta r_{\parallel}$  and  $\Delta r_{\perp}$ have opposite sign and are independent of the constants c, which are so difficult to deduce.

This behavior is seen qualitatively in Figs. 1(a) and 1(b), and in Fig. 2 we compare the steepness of the observed length changes with the behavior predicted. If  $\Delta r/rH_i\Delta M$  is plotted against  $1-\mu$ , a straight line is found. Its steepness and intercept of the ordinate give the elastic constants. Thus our experiments provide a somewhat round-



FIG. 2. Steepness of the small reversible curves indicated by pairs of arrows in Figs. 1(a) and 1(b). The full lines represent calculations and the points, measurements.

about but nevertheless feasible technique for measuring the elastic constants.

Table I shows elastic constants deduced from our magnetostriction measurements compared with direct observations. There is a good agreement between values in the literature for the pure metals and the observed values for the Ta-5at.% Nb and the In-9at.\% Pb samples. However, Young's modulus in In-14at.% Tl differs from the value for pure indium by a factor of 3. A direct measurement of Young's modulus in this alloy was carried out to check this result, and fair agreement was found.

We have imagined our sample to consist of a core of reversible type-II superconductive material and a surface capable of carrying superconductive screening currents, when it is in the mixed state. This model can be extended in a simple way to take into account pinning forces in the core of the sample also. In their presence gradients in the density of the flux lines are easily built up by any change of the external field. The gradients are equivalent to a macroscopic current density within the specimen.<sup>8</sup> For such currents Eqs. (1) are valid too. Also Eqs. (2) can be used for internal currents because  $\Delta M$ weights the distorted volume correctly for a first approximation.

Figure 1(b) shows a well-defined dip just below  $H_{C2}$ . The magnetization curve has a rather steep decay at this point. We suppose that near this field the surface and the interior lose their capacity for carrying a screening current, so that there is a sudden entry of flux into the specimen, and the forces which have distorted the sample before relax. The sample becomes thicker and shorter. This is shown in Fig. 1(b) by the rise from the deepest point and in Fig. 1(a) by the very steep decay in increasing field near  $H_{C2}$ .

The flat ellipsoid used in this work has one disadvantage for the measurement of magnetostriction due to surface currents. If the equatorial plane of the sample does not lie exactly parallel to the external magnetic field, a large magnetic moment is induced at an angle to  $H_{\rho}$  and therefore gives rise to forces distorting the sample. We have chosen a flat ellipsoid for our experiment because we were mainly interested in the superconducting properties, for which a small demagnetization factor is desired. The angle of the external field has been chosen so as to give the expected magnetostriction curve in the Meissner state for a flat ellipsoid with an axial ratio of 1:10. Thus this part of the curve includes no experimental information.

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Table I. Comparison of elastic constants deduced from this work with values from direct measurements.  $\sigma$  is the Poisson number and E is Young's modulus in units of 10<sup>9</sup> dyn/cm<sup>2</sup>.

Elastic constants derived from				
Magnetostriction measurements		Direct measurements		
$\sigma = 0.345 \pm 0.015$	$E = 2120 \pm 100$	$\sigma = 0.340$	<i>E</i> = 1860	
Ta-5 at.% Nb, single crystal, 2.6°K;		Pure Ta, <sup>a</sup> 293°K.		
cylinder: axial ratio 1:20.				
$\sigma = 0.47 \pm 0.02$	$E = 204 \pm 15$	$\sigma = 0425$	E = 207	
In-9 at.% Pb, polycrystalline, 2.4°K;		Pure In, <sup>a</sup> 0°K.		
long ellipsoid: a	xial ratio 1:8.			
$\sigma = 0.47 \pm 0.03$	$E = 70 \pm 30$	$E = 115 \pm 30$		
In-14 at.% Tl, polycrystalline, 1.3°K;		Direct measurement on a rod		
flat ellipsoid: axial ratio 1:10.		of In-14 at.% Tl at 4.2°K.		

<sup>a</sup>O. L. Anderson, in <u>Physical Acoustics</u>, edited by W. P. Mason (Academic Press, Inc., New York, 1965), Vol. III, pt. B, pp. 87, 88.

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<sup>1</sup>D. G. Schweitzer, Phys. Letters <u>28A</u>, 98 (1968).

<sup>2</sup>G. K. White, Cryogen. <u>1</u>, 151 (1961).

<sup>3</sup>G. Brändli and F. D. Enck, Phys. Letters <u>26A</u>, 360 (1968).

<sup>4</sup>G. Brändli, F. D. Enck, E. Fischer, and J. L. Olsen, Helv. Phys. Acta 41, 706 (1968).

<sup>5</sup>P. S. Swartz and H. R. Hart, Jr., Phys. Rev. <u>137</u>, A818 (1965).

<sup>6</sup>H. J. Fink, Phys. Rev. Letters <u>14</u>, 309 (1965).

<sup>7</sup>J. G. Park, Phys. Rev. Letters <u>15</u>, 352 (1965).

<sup>8</sup>C. P. Bean, Rev. Mod. Phys. <u>36</u>, 31 (1964).

## SMALL-ANGLE X-RAY SCATTERING OF QUARTZ NEAR THE $\alpha$ - $\beta$ TRANSITION

H. Brumberger, W. Claffey, N. G. Alexandropoulos, and D. Hakim Department of Chemistry, Syracuse University, Syracuse, New York 13210 (Received 29 January 1969)

A series of experiments on the small-angle x-ray scattering of quartz over a temperature range centered on the  $\alpha$ - $\beta$  transition (573°C) showed no excess scattering during the phase transformation. This is taken to indicate that the previously reported "opalescence" (Yakovlev, Velichkina, and Mikheeva and Yakovlev and Velichkina) is probably not a critical phenomenon but arises from twinning inhomogeneities.

Numerous papers dealing with various aspects of the  $\alpha$ - $\beta$  quartz transition at 573°C have been published (for a review, see Young<sup>1</sup>); there still, however, appears to be some uncertainty concerning the order of the transition. Various measurements<sup>1,2</sup> tend to support the view that this is a first-order transformation. Nevertheless, an enormous light-scattering effect, akin to critical opalescence, has been reported<sup>3</sup>; the intensity of light scattered at an angle of 90° apparently increases by a factor of  $10^4$  near the transition temperature relative to the intensity scattered at 20°C. This phenomenon is ascribed to fluctuations in the refractive index due to corresponding fluctuations in a characteristic Landau order parameter  $\eta$ .<sup>4-6</sup> According to Landau,<sup>7</sup> an increase in the diffuse scattering of x rays from order fluctuations should also be observed near the Curie point. Recently, the Raman and Brillouin spectra of quartz near  $T_{\alpha\beta}$  have been studied.<sup>8</sup> While "opalescence" was indeed observed, its origin was assigned to scattering by domain walls between electrical twins rather than to critical fluctuations in the order parameter.

We have examined the small-angle x-ray scattering of quartz to determine whether sufficiently large electron-density fluctuations to cause an observable increase in scattered intensity were indeed present near  $T_{\alpha\beta}$ . If there are critical fluctuations in the order parameter (i.e., displacements of Si atoms from  $\beta$ -phase equilibrium positions) local fluctuations in electron density would be expected, as well as increasingly longrange correlation of such fluctuations as the transition is approached.

The scattering of 1-mm thick quartz plates of several origins<sup>9</sup> was measured in transmission with a Kratky camera,<sup>10</sup> using Cu  $K\alpha$  or Mo  $K\alpha$ radiation monochromatized by the appropriate  $\beta$ filter and pulse-height discrimination. The samples were confined in a furnace described elsewhere<sup>11</sup>; temperature constancy over the illuminated portion of the sample could be controlled to ±0.05°C. Iron-Constantan thermocouples, calibrated against National Bureau of Standardscertified thermometers, were used to measure the temperature; the occurrence of the transitions at 573°C was checked by observing the change of intensity of the 100 line with temperature (using the same thermocouple) in a diffractometer.<sup>1</sup>

Three types of experiments were performed. Scattering curves as a function of angle were obtained at a series of temperatures between 567 and 575°C (including 571.0 and 574.0°C) and at 195°C. Within experimental error these proved to be the same. In another series, the intensity at several fixed angles was measured in steps of 3°C between 535 and 595°C, again showing no significant change in intensity at any temperature or angle. Since it is possible that the "opalescence" effect exists in an extremely narrow temperature range near  $T_{\alpha\beta}$ , a series of experiments were