## Acoustic Attenuation Due to Domain Walls in Anisotropic Superconductors, with Application to $U_{1-x}Th_xBe_{13}$

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We propose an explanation of the large sound-attenuation peak observed at the lower transition,  $T_{c2}$ , in superconducting  $U_{1-x}Th_xBe_{13}$ . It assumes that the transition is between two different anisotropic superconducting phases, and that the low-temperature phase is tetragonally distorted. The domain-wall energy is very small at  $T_{c2}$  and the attenuation due to the motion of walls is very large. This attenuation depends strongly on direction and polarization, and so the model may be easily tested. The implications for the phase diagram of these alloys are discussed.

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In a recent Letter Batlogg et al.<sup>1</sup> found that the ultrasonic attenuation in the superconducting phases of  $U_{1-x}Th_xBe_{13}$  (x = 0.0175) was very strong with a sharp maximum at  $T = T_{c2}$ —the transition temperature of the second transition discovered in specificheat measurements by Ott et al.<sup>2</sup> Batlogg et al.<sup>1</sup> proposed that this very strong attenuation was due to an additional magnetic order although from NMR measurements and other considerations the ordered moment must be very small (  $\leq 10^{-2} \mu_{\rm B}/{\rm U}$ ).<sup>3</sup> In this Letter we wish to propose an alternative mechanism to explain the observations of Batlogg et al.,<sup>1</sup> namely that because of twinning there exist domain walls in the sample between differently oriented anisotropic superconducting domains which can move and damp the sound wave. Joynt and Rice<sup>4</sup> (JR) pointed out that in a cubic system the anisotropic superconducting state drives a tetragonal or rhombohedral lattice distortion. Therefore, there can be a natural coupling of a sound wave to the domain walls leading to a large increase in attenuation and a softening of the elastic constants. This effect is seen in other phase transitions in which there is a secondary lattice distortion.<sup>5,6</sup> We make specific predictions for the attenuation in different directions and polarizations from this mechanism. Verification of these predictions would be an unambiguous proof of anisotropic and therefore unconventional superconductivity. Although it would not completely determine the symmetry of the order parameter, the range of possible states would be very much narrowed.

We begin by making some general proposals for the

phase diagram of  $U_{1-x}Th_xBe_{13}$ .<sup>2,7</sup> Since the second phase transition is only observed for  $x \ge 0.0175$  and since there are indications of a dip in the onset  $T_c(x)$ at  $x \approx 0.015$ , we are led to propose a phase diagram with three superconducting phases which we label A, B, and C (see Fig. 1). The following remarks can be made. (a) Since  $T_c^A(x) \neq T_c^B(x)$ , A and B must belong to different representations chosen from the  $A_1$ , E,  $T_1$ , or  $T_2$  representations. (A number of authors have listed the possible superconducting states in a cubic crystal with spin-orbit coupling.<sup>8–11</sup> We will adopt the identifying serial numbers given by Blount.<sup>11</sup>) (b) The B-C transition presumably takes place between phases belonging to the same representation especially since experiments show it is not strongly first order. (c) The very different acoustic attenuation properties of samples<sup>12</sup> with x = 0.0175 and x = 0 is consistent with this proposal that A and C are different phases.

To explain the acoustic attenuation data we propose that phase C is one of the tetragonally distorted phases found by JR. Such a phase has either three or six degenerate ground states. In the presence of the (100) longitudinal sound wave of the experiment, domains oriented along the direction of polarization will have an instantaneously different free energy from those oriented perpendicularly. Thus the sound wave induces an oscillation of walls separating these domains. The damping of this motion leads to acoustic attenuation.

The specific model we consider is that of a domain wall pinned at random locations separated, on average, by a distance  $L_i$ .<sup>13</sup>



FIG. 1. Proposed phase diagram of  $U_{1-x}Th_xBe_{13}$  in the temperature-impurity-concentration plane. A, B, and C are different anisotropic superconducting phases. The two main possibilities (i) and (ii) consistent with our model are indicated explicitly. (ii) has two subpossibilites because the  $T_1$  and  $T_2$  representations cannot be distinguished by sound experiments. Circles are data from Ref. 7, crosses from Ref. 2. Dashed lines indicate the proposed separation of phases. The region  $x > 5 \times 10^{-2}$  contains only a single experimental point—the lines drawn there are purely conjectural. The arrows indicate temperatures above which there is definitely only one transition (H.R. Ott, private communication). The notation for the phases, and the justification of the possibilities given, are explained in the text.

There are two possibilities for the damping mechanism, which we consider in turn.

Viscous damping.—The equation of motion for the lateral coordinate z of a wall in the x-y plane is

$$0 = f \nabla^2 z(x, y) - \Gamma \dot{z} + \gamma_x e^{i\omega t} (b_1 - b_2).$$
 (1)

Here f is the wall energy per unit area,  $\Gamma$  is the friction coefficient,  $\gamma_x$  is the maximum value of the traceless strain  $\gamma_x = [a_x - (a_y + a_z)/2]/a$ , the a's denoting lattice constants, and  $b_1$ ,  $b_2$  are the free energy densities per unit strain of domains oriented in the x, y (or z) directions, respectively.  $\omega$  is the acoustic frequency. The attenuation from the lowest mode of the wall can now be calculated if we assume that the wavelength is much greater than  $r_0$ , the average radius of a domain. It is

$$\alpha_{s} = 4 \frac{(b_{1} - b_{2})^{2}}{\Gamma r_{0} \rho c_{s}^{3}} \frac{\omega^{2}}{\omega^{2} + \omega_{p}^{2}},$$
(2)

where  $\omega_p = 2\pi^2 f / L_i^2 \Gamma$ ,  $\rho$  is the mass density, and  $c_s$  is



FIG. 2. The frequency dependence of the attenuation at  $T = T_{c2}$ . The points are data from Ref. 1. The curve is the theoretical form for viscous damping:  $\alpha_s = \alpha_0 \omega^2 / (\omega^2 + \omega_p^2)$ , with  $\alpha_0 = 0.266$  dB/ $\mu$ sec and  $\omega_p = 90.0$  MHz. The fit is better than the linear form proposed in Ref. 1, but more experimental work is clearly needed.

the sound velocity. A fit of the equation to the frequency data of Batlogg *et al.*<sup>1</sup> is given in Fig. 2. Note that for  $\omega \ll \omega_p$ ,  $\alpha_s \sim f^{-2}$ , but for  $\omega \gg \omega_p$ ,  $\alpha_s$  is independent of f.

Hysteretic damping.—In this case we take  $z_0(x,y,t)$ from the solution of Eq. (1) with  $\Gamma = 0$ , but with the actual displacement  $z(x,y,t) = z_0 \pm L_h^{-1}(z_0^2 - z_m^2)$ , where the upper (lower) sign is to be taken when  $\dot{z} > 0$  ( $\dot{z} < 0$ ).  $z_m(x,y)$  is the maximum displacement at (x,y) and  $L_h^{-1}$  is the shape parameter for the hysteresis loop, roughly interpretable as the number of traps per unit displacement met by the wall. Then one finds an attenuation

$$\alpha_{s} \sim \frac{\gamma_{x}^{0} |b_{1} - b_{2}|^{3} L_{i}^{4} L_{h}^{-1}}{r_{0} \rho c_{s}^{3} f^{2}} \omega, \qquad (3)$$

for small  $\omega$ .  $\gamma_x^0$  is the strain amplitude. The linear dependence on frequency is also consistent with the data of Fig. 2. It is clear that further experimental work on the frequency and amplitude dependence can decide between the two damping mechanisms. The expressions (2) and (3) are consistent with earlier work on a somewhat different model for magnetic domains.<sup>6</sup>

The main point of this Letter concerns the dependence on f, the wall energy. Near the first-order transition into a tetragonal state f vanishes, or nearly so. This gives the very large peak in  $\alpha$  observed at  $T_{c2}$ . The vanishing of f comes about as follows: The generic form of the free energy for a three-dimensional representation is

F

$$= -\alpha |\mathbf{u}|^{2} + \beta (\mathbf{u} \cdot \mathbf{u}^{*})^{2} + \delta_{1} |\mathbf{u}^{2}|^{2} + \delta_{2} \sum_{i} |u_{i}|^{4} + \delta_{3} \times (\text{gradient terms}). \quad (4)$$

The  $u_i$  are the components of the order parameter d:  $\mathbf{d} = \sum_i u_i \mathbf{d}_i$ , where the  $\mathbf{d}_i$  are a basis for the representation. The gap matrix is  $\Delta = i \sigma_y \boldsymbol{\sigma} \cdot \mathbf{d}$ , the  $\sigma$ 's being the Pauli matrices. The stability of phases in the representation is determined by the signs of  $\delta_2$  and  $2\delta_1 + \delta_2$ . Following Volovik and Gorkov,<sup>9</sup> the domain-wall solutions may be found. The wall energy per unit area has the form

$$f \sim \alpha^{3/2} (\beta + \delta_1)^{-3/2} \delta_2^{1/2} |\delta_3|^{1/2}.$$

We assume that the lower transition is driven by a change in sign of the anisotropy parameter  $\delta_2$ . Then the transition is weakly first order and the wall energy vanishes to fourth order:  $f \sim t_2^{1/2}$  + (small corrections), where  $t_2 = 1 - T/T_{c_2}$ . Since, for hysteretic damping, or viscous damping for  $\omega < \omega_p$ ,  $\alpha_s \sim f^{-2}$ , we expect  $\alpha_s \sim t_2^{-1}$  with rounding of the singularity at small  $t_2$ . We have also calculated the fractional change in sound velocity and find  $\Delta c_s/c_s \sim -f^{-1} \sim -t_2^{-1/2}$ . This is also in agreement with the data of Ref. 1. The experimental width of the transition is too large to allow a quantitative comparison between theory and experiment, as far as the temperature dependences are concerned.

Nevertheless, there is a very clear-cut test of the present model which may be carried out. It was pointed out above that the free energies of the various domains depend on the local strains produced by the (100) longitudinal wave. Other polarizations and directions will couple with different strengths or not at all. These couplings, as defined by Eq. (1), can be calculated with the results of JR. Coupling strengths of domains for the most commonly used sound waves are given in Table I. More difficult to compute are the couplings of the rhombohedral phases, but these are expected to be much smaller. So only the presence or

absence of coupling is given for these.

What is important is that for certain sound waves, notably transverse (100) and longitudinal (111), there is *no* coupling to the tetragonal phase. Therefore, these waves should *not* show the large attenuation peak below  $T_{c2}$  observed for the longitudinal (100). This characteristic sharply differentiates the concept of a transition between different superconducting phases from the hypothesis of low-temperature antiferromagnetism. We propose this strong anisotropy of the attenuation as a stringent test of the putative phase diagram (Fig. 1).

We turn now to the consequences for the phase diagram with phases A, B, and C assuming that the proposal that the C phase has a tetragonal distortion is correct. Two possibilities can be distinguished:

(i) The C phase belongs to the 2D E representation, i.e., either of phases 3 or 4. In this case the B phase must be phase 5 and is cubic and the A phase presumably is one of phases 10, 12, or 13 belonging to  $T_1$  or  $T_2$  since there is no strong attenuation of (100) sound waves and it has point zeros.<sup>14</sup>

(ii) The C phase belongs to the 3D  $T_1$  or  $T_2$  representation, i.e., one of the phases 6–9. In this case the B phase must be rhombohedrally distorted, i.e., one of phases 10–13, and the A phase can belong to the *E* representation (phases 4, 5) or a rhombohedrally distorted phase from the other member of  $\{T_1, T_2\}$ . It is not clear which of (i) or (ii) is correct. An argument in favor of (i) is that a phase transition from phase 3 or 4 to 5 is easy to rationalize since phase 5 has many more (eight) zeros in the energy gap and thus would have the larger entropy. On the other hand (ii) offers a natural explanation of the absence of anomalous attenuation in A, since A can be the cubic phase 5. Also we note that in this case C could be one

TABLE I. The free energy per unit strain, as a fraction ot total condensation free energy, of minority  $(b_1)$  and majority  $(b_2)$  domains is given in the form  $(b_1, b_2)$  for various sound waves. Standard notation is employed for the latter. The attenuation depends on  $|b_1 - b_2|$ . For the (110) T<sub>2</sub> wave there are three inequivalent domains. In the last row the presence or absence of a small coupling to rhombohedrally distorted phases is indicated.

| Direction | Polarization   | Phase   |  |  |  |                   |
|-----------|----------------|---|--|--|--|-------------------|
|           |                | 3   | 4  | 6 or 7   | 8 or 9   | 10, 11, 12, or 13 |
| (100)     | L              | $\left(-\frac{1}{3},\frac{1}{6}\right)$       | $\left(\frac{1}{3},-\frac{1}{6}\right)$        | $(\frac{1}{3},-\frac{1}{6})$                   | $\left(-\frac{1}{6},\frac{1}{12}\right)$       | None              |
|           | Т              | (0,0)   | (0,0)  | (0,0)  | (0,0)  | None              |
| (110)     | L              | $(\frac{1}{6}, -\frac{1}{12})$                | $(-\frac{1}{6},\frac{1}{12})$                  | $(-\frac{1}{6},\frac{1}{12})$                  | $(\frac{1}{12}, -\frac{1}{24})$                | Small             |
|           | T <sub>1</sub> | (0,0)   | (0,0)  | (0,0)  | (0,0)  | None              |
|           | T <sub>2</sub> | $(-\frac{1}{12},\frac{1}{4},0)$               | $(\frac{1}{12}, -\frac{1}{4}, 0)$              | $(\frac{1}{12}, -\frac{1}{4}, 0)$              | $(-\frac{1}{24},\frac{1}{8},0)$                | None              |
| (111)     | L              | (0,0)   | (0,0)  | (0,0)  | (0,0)  | Small             |
|           | Т              | $(-\frac{1}{6}\sqrt{2},\frac{1}{12}\sqrt{2})$ | $(\frac{1}{6}\sqrt{2}, -\frac{1}{12}\sqrt{2})$ | $(\frac{1}{6}\sqrt{2}, -\frac{1}{12}\sqrt{2})$ | $(-\frac{1}{12}\sqrt{2},\frac{1}{24}\sqrt{2})$ | Small             |

of the weakly ferromagnetic phases in which case we would expect many domain walls as pointed out by Volovik and Gorkov.<sup>9</sup> Two final comments: We have made our discussion only in terms of odd-parity states but, as is clear from the work of Volovik and Gorkov,<sup>9</sup> there are similar even-parity states which cannot be distinguished in these experiments; also, we cannot rule out the possibility that the B phase is a glassy superconducting phase, as proposed by Volovik and Khmel'nitskii.<sup>15</sup>

In conclusion, we have shown how the additional attenuation due to domain-wall motion may explain the anomalous attenuation observed in  $U_{1-x}Th_xBe_{13}$  with x = 0.0175. Experimental tests of our proposal have been suggested. Verification of our proposal would be an unambiguous proof that the C phase is anisotropic and therefore an unconventional superconductor.

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