small peak at a low positive field is the previously mentioned dielectric anomaly. The position of the hole resonance calculated numerically from (3) has been found to be in close agreement with that given by (1).

It has been shown that Eq. (1) describes both a Doppler-shifted cyclotron resonance condition and the onset of strong damping of the Alfvén waves in bismuth, corresponding to real transitions between Landau levels induced by the electromagnetic field of wave vector  $k_A$ . Undamped electromagnetic waves ("helicons") also occur in charged plasmas, <sup>6,7</sup> and the damping criterion is given by a similar relation.<sup>2</sup>

The authors thank A. G. Redfield for valuable suggestions throughout the course of this work. One of us (J.K.) would also like to thank him for his encouragement and sponsorship of this work as part of a Columbia University thesis. The authors also thank Y. H. Kao for providing several bismuth crystals and R. R. Haering for helpful discussions.

- <sup>1</sup>S. J. Buchsbaum and J. K. Galt, Phys. Fluids  $\underline{4}$ , 1514 (1961).
- <sup>2</sup>P. B. Miller and R. R. Haering, Phys. Rev. <u>128</u>, 126 (1962).
- $^{3}$ J. K. Galt, W. A. Yager, F. R. Merritt, B. B. Cetlin, and A. D. Brailsford, Phys. Rev. <u>114</u>, 1396 (1959).
- <sup>4</sup>A. L. Jain and S. H. Koenig, Phys. Rev. <u>127</u>, 442 (1962).

<sup>5</sup>J. Kirsch (to be published).

- <sup>6</sup>P. Aigrain, <u>Proceedings of the International Con-</u> ference on Semiconductor Physics, Prague, 1960
- (Czechoslovakian Academy of Sciences, Prague, 1961). <sup>7</sup>F. E. Rose, M. T. Taylor, and R. Bowers, Phys. Rev. 127, 1122 (1962).

## MAGNETIZATION OF V<sub>3</sub>Si THIN FILMS

## J. J. Hauser

Bell Telephone Laboratories, Murray Hill, New Jersey (Received October 4, 1962)

The magnetization of hard superconductors can be described in terms of two models: the model developed by Bean<sup>1</sup> which is based on a shielding of the sample by filaments and the model based on negative surface energy.<sup>2,3</sup> The essential difference in the two models lies in the fact that the former predicts a size-dependent magnetization behavior and the latter is reversible. This effect was first shown by Bean<sup>1</sup> who used two cylinders of Nb<sub>3</sub>Sn of different diameters and by Williams<sup>4</sup> on V<sub>3</sub>Ga powders. We have examined the magnetization of  $V_3Si$  as a function of sample size over a range from  $5 \times 10^{-5}$  to 0.1 cm and also as a function of temperature. These experiments demonstrate that neither the filamentary model nor the negative surface energy model provides by itself an adequate description of the behavior. The results suggest that a combination of the features of both models is required.

The Bean model predicts that the sample will remain perfectly diamagnetic up to the thermodynamic critical field, and the shielding thereafter will be performed by the filaments in a manner characterized by the parameter  $H^* = (4\pi/10)RJ_c$ , where R is the radius of a cylinder or half the thickness of a plate, and  $J_c$  the macroscopic critical current density of the sample. If  $H^*$  could

be made very small compared with  $H_c$ , one should then approach the magnetization curve of a soft superconductor with bulk critical field  $H_c$ . There are two different ways to make  $H^*$  small: One is to obtain a thin film of the material, the other is to measure the sample at a temperature close to  $T_c$  so that  $J_c$  is small. Using a vibrating-sample magnetometer, the size dependence has been investigated by measuring plates ranging in thickness from 1 mm to 47000 Å, and the results are shown in Figs. 1 and 2. The temperature dependence was studied by measuring the samples at two temperatures: 4.2°K in Fig. 1 and 14.25°K in Fig. 2. The thermodynamic critical field at 0°K calculated from the specific heat measurement of Morin and Maita<sup>5</sup> is 6370 gauss. The critical field at 14.25°K using a parabolic dependence of temperature is 1785 gauss. It is evident from Figs. 1 and 2 that the deviation from linearity in the magnetization, especially in the thinner samples, occurs at fields well below the thermodynamic critical field. Furthermore, the thin films exhibit a critical slope smaller than one, which indicates that these films are partially penetrated by the magnetic field.

The field penetration at values smaller than the thermodynamic critical field could be caused by



FIG. 1. Magnetization per unit volume as a function of applied magnetic field for  $V_3Si$  samples of various thicknesses at  $4.2^{\circ}K$ .

four effects: (1) a geometrical intermediate state caused by the shape and orientation of the sample, (2) a mixed state occurring because of the presence of a second phase with a lower critical field, (3) a mixed state produced by the negative surface energy conditions existing in the material, and (4) a penetration depth large compared with the size of the sample. All the samples investigated were approximately 1 cm  $\times$  0.5 cm and their thicknesses are reported in Figs. 1 and 2. A lead foil 250 000A thick, and a thin lead film 20 000Å thick, both 1 cm  $\times 0.5$  cm, were measured, and a nearly ideal superconducting behavior was observed in both cases. In the case of the foil, the transition from superconducting to normal occurred within 5 gauss, the magnetization was reversible without hysteresis, and the critical field  $H_0$  was 817 gauss which is very close to the value reported in the literature. As the other samples were lined up as accurately with respect to the magnetic field, this experiment rules out the possibility of an orientation-induced intermediate state. The V<sub>3</sub>Si single crystal was prepared by E.S. Greiner by the floating zone technique and no second phase was detected by optical examination at 300× magnification. As shown in Figs. 1 and 2, the single crystal yields approximately the same magnetization curve as a cast polycrystalline sample of approximately the same size. Consequently, the observed field penetration cannot be explained by the presence of a second phase. The thin films which were prepared by the reduction of mixed chlorides<sup>6</sup> yield data which are consistent with that obtained on bulk. Indeed, the thinnest bulk piece measured is  $3 \times 10^{-2}$  cm and the thickest film investigated is  $6 \times 10^{-3}$  cm, which are less than an order of magnitude apart.



FIG. 2. Magnetization per unit volume as a function of applied magnetic field for  $V_3Si$  samples of various thicknesses at 14.25°K.

Consequently, the field penetration which occurs at values much smaller than the bulk thermodynamic critical field must be due to negative surface energy.

Abrikosov<sup>2</sup> as well as Goodman<sup>3</sup> have shown that when the mean free path in a material becomes very small, the surface energy between normal and superconducting regions becomes negative. In this case, the material spontaneously splits up into a laminar structure at a field  $H_{c_1}$ , smaller than  $H_c$ , and becomes normal at an upper transition field  $H_{c_{2}}$  larger than  $H_{c}$ . This magnetization curve should, however, be reversible and the samples should show a Meissner effect for fields smaller than  $H_c$ . In samples thicker than 500 000 Å, the flux trapped by reducing the field from 18 kG to zero is approximately equal to the maximum field that the sample can shield. This is to be expected in such samples where the filamentary shielding is so large. However, even the thinnest film measured (8000 Å) showed hysteresis and flux trapping. It is true, however, that in this thin film the trapped flux was much smaller than the maximum shielded field. This departure from an ideal negative surface energy behavior can be explained as follows: As soon as spontaneous splitting up into reversible filaments occurs at  $H_{c_1}$ , the superconducting filaments get pinned by existing lattice defects such as dislocations and, consequently, this leads to irreversibility. Another discrepancy with the negative surface energy model is the lack of Meissner effect even for fields smaller than  $H_c$ . Even the thinnest samples will have filaments and, as is well known, the dependence of the filamentary transition field on temperature is close

to linear. The dependence of the thermodynamic critical field on temperature is, on the other hand, close to parabolic. Consequently, if one cools a sample under an applied magnetic field, no matter how small, the filaments will become superconducting first, and will trap the field and thereby prevent the occurrence of a full Meissner ef fect.7

The data presented here can, therefore, be interpreted in terms of the two models: The basic magnetization curve is the one predicted by the negative surface energy model which leads to field penetration at values smaller than  $H_c$ ; as the sample becomes larger this basic curve becomes obscured by the shielding of filaments rigidly fixed in the material.

The deviation of the initial slope of magnetization versus field from  $-1/4\pi$  in the thinner films can be explained if one assumes that the film is partially penetrated by the magnetic field. If one fits the slopes of various films to the London equation,

$$-4\pi M/H = 1 - (2\lambda/d) \tanh(d/2\lambda) \tag{1}$$

where M is the magnetization per unit volume;  $\lambda$ , the penetration depth; and d, the thickness of the film. The average penetration depth measured at low fields was 15000 Å at  $4.2^{\circ}$ K. This extremely large penetration depth is consistent with the predictions made by Clogston and coworkers<sup>8</sup> on the  $\beta$ -wolfram superconductors from their nuclear magnetic resonance (nmr) measurements. This result is also in fair agreement with an independent measurement made by Greytack.<sup>9</sup> A preliminary measurement on one film would indicate that  $\lambda (14.25^{\circ} \text{K}) / \lambda (4.2^{\circ} \text{K}) = 2.45$ . If one calculates the same ratio using the well known empirical formula<sup>10</sup>

$$\lambda(T)/\lambda(0) = [1 - (T/T_c)^4]^{-1/2},$$
 (2)

one finds  $\lambda (14.25^{\circ} \text{K}) / \lambda (4.2^{\circ} \text{K}) = 1.45$ . Consequently,

the variation of the penetration depth with temperature in V<sub>3</sub>Si is much more rapid than the usually observed variations.

In conclusion, these experiments show again the validity of the filamentary model proposed by Bean to explain the magnetization of hard superconductors. However, once the size dependence is eliminated by measuring the thinner samples the underlying curve is that proposed by the negative surface energy model. Actually, it is more than likely that both models apply in a real material, and that negative surface energy is what is required to make a defect such as a dislocation behave as a filament.<sup>11</sup> A penetration depth of approximately 15000 Å was measured in the thinner films and was found to increase with temperature faster than the usually observed dependence.

It is a pleasure at this time to thank H. C. Theuerer and D. D. Bacon for producing the thin films, E. S. Greiner for supplying the  $V_3$ Si single crystal, D. Dorsi for making and machining the polycrystalline V<sub>3</sub>Si samples, and W. H. Haemmerle for his help in obtaining the data.

<sup>4</sup>H. J. Williams (private communication).

<sup>6</sup>J. J. Hauser and H. C. Theuerer, Phys. Rev. (to be published).

<sup>7</sup>F. J. Morin, J. P. Maita, H. J. Williams, R. C. Sherwood, J. H. Wernick, and J. E. Kunzler, Phys. Rev. Letters 8, 275 (1962).

<sup>8</sup>A. M. Clogston, A. C. Gossard, V. Jaccarino, and Y. Yafet, Phys. Rev. Letters 9, 262 (1962).

<sup>9</sup>T. J. Greytack (private communication).

<sup>10</sup>C. J. Gorter and H. B. G. Casimir, Physik. Z.  $\frac{35}{11}$ , 963 (1934). <sup>11</sup>C. J. Gorter, Phys. Letters <u>1</u>, 69 (1962).

<sup>&</sup>lt;sup>1</sup>C. P. Bean, Phys. Rev. Letters 8, 250 (1962).

<sup>&</sup>lt;sup>2</sup>A. A. Abrikosov, J. Exptl. Theoret. Phys.

<sup>(</sup>U.S.S.R.) 32, 1442 (1957) [translation: Soviet Physics-JETP 5, 1174 (1957)].

<sup>&</sup>lt;sup>3</sup>B. B. Goodman, Phys. Rev. Letters 6, 597 (1961); IBM J. Research. Develop. 6, 63 (1962).

<sup>&</sup>lt;sup>5</sup>F. J. Morin and J. P. Maita (to be published).