HEAT CAPACITY EVIDENCE FOR A LARGE DEGREE OF SUPERCONDUCTIVITY IN V_3Ga IN HIGH MAGNETIC FIELDS

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It has generally been assumed that for materials in which superconductivity persists in fields above the thermodynamic critical field, there is only a small fraction of the volume involved which is associated with superconducting filaments.[1,2] However, interpretation of the heat capacity of V_3Ga, which we have measured in fields as large as 70 kgauss, leads us to the opposite conclusion. These data suggest that most of the material remains superconducting even at fields many times the thermodynamic critical field, in spite of the fact that magnetic susceptibility data show that the magnetic field has penetrated most of the sample.

Heat capacity measurements were made in an apparatus utilizing pulse techniques which will be described elsewhere.[3] The measurements were made on a 3-gram cubical sample of polycrystalline V_3Ga over the temperature range 25°K to 1.5°K in fields of 0, 18, 40, and 70 kgauss. Heat capacity curves, including one showing some experimental points, are shown in Fig. 1. Normally, the measurements were made by cooling the sample from a temperature above 25°K in a magnetic field, and the data taken as the temperature was decreased. However, in experiments at both 14 and 70 kgauss, observations were also made as the temperature was increased. In addition, the sample was cooled in the absence of a magnetic field to 11°K. A magnetic field of 14 kgauss was then applied, and data were taken with increasing temperature followed by decreasing temperature over the interval 11 to 20°K. Within the accuracy of the measurements, the data agreed in all cases; no evidence of hysteresis was observed.

The heat capacity in the normal state, based on the data taken at temperatures above 15°K, is represented by

$$C_p = 244\times10^{-4}T + 6.5\times10^{-5}T^3 \text{ cal mole}^{-1}\,°\text{K}^{-1}. \qquad (1)$$

So far as we know, this is the largest linear term, or γ, thus far reported for any substance. It is consistent with susceptibility data in the normal state and the narrow-band model proposed by Clogston and Jaccarino.[4] The total entropy at the critical temperature, 14.66°K, is 0.43 cal mole^{-1} °K^{-1}, and to the extent that Eq. (1) is valid, it can be separated as follows:

$$S_{(\text{total, } 14.66°\text{K})} = 0.43 \text{ cal mole}^{-1}\,°\text{K}^{-1}, \qquad (2)$$

$$S_{(\text{lattice, } 14.66°\text{K})} = 0.07 \text{ cal mole}^{-1}\,°\text{K}^{-1}; \qquad (3)$$

$$S_{(\text{electronic, } 14.66°\text{K})} = 0.36 \text{ cal mole}^{-1}\,°\text{K}^{-1}. \qquad (4)$$

It is significant that the total entropy at 14.66°K, as determined from each series of data in differing fields, yields the same value to within 1%. This rules out the possibility that a latent heat was present but somehow missed, unless the absence of an observable latent heat was due to inhomogeneity of the sample. However, it seems

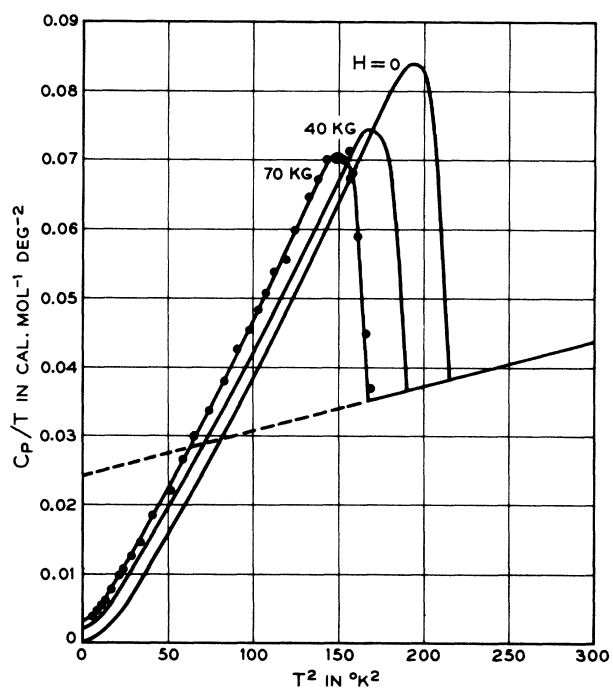

FIG. 1. The heat capacity of V_3Ga in fields of 0, 40, 70 kgauss. The dashed line represents the extrapolated behavior expected for the normal state based on measurements above the critical temperature (14.66°K). The persistence of the heat capacity peak in the magnetic field and the smallness of the intercept at 0°K compared with γ (straight line) are evidence for a large degree of superconductivity in high magnetic fields.

Table I. Parameters which indicate the rate of transition of V_3Ga from the superconducting to normal state with applied magnetic field.

Applied field (kgauss)	Transition temperature (°K)	Entropy of peak (above normal state) (cal mole^{-1} °K^{-1})	Intercept of C_p/T at 0°K (cal mole^{-1} °K^{-2} $\times 10^4$) ($\gamma = 244$)
0	14.66	0.135	0
18	14.28	0.125	9
40	13.81	0.108	19
70	12.92	0.097	32

more likely that a latent heat does not exist in this type of material.[5]

The most surprising features of the data are the persistence of the heat capacity peak in a high magnetic field and the small change in transition temperature with field.[6] The change in transition temperature (Column 2, Table I) with magnetic field leads to a critical field value of 600 kgauss at 0°K using a linear extrapolation (which is expected to be too high, since the third law of thermodynamics requires a zero slope at 0°K) and a value of about 300 kgauss using a parabolic extrapolation. These values are consistent with the analogous values, 800 kgauss and 400 kgauss, obtained from electrical data[7] (using low current densities) which are expected to be somewhat higher by virtue of the different nature of the measurements. A value of 6000 gauss is obtained for the critical field of the bulk material at 0°K by using a value of 244 $\times 10^{-4}$ cal mole^{-1} for γ and BCS theory.[8]

A measure of the persistence of the heat capacity peak with field is given by the amount of entropy associated with the region having a heat capacity larger than that of the normal state (i.e., the area above the linear curve). These values are shown in the third column of Table I. These data suggest that most of the sample still remains superconducting at 70 kgauss. The values of the intercepts at 0°K of C_p/T curves (Column 4, Table I), compared with the value of γ (244×10^{-4} cal/mole), also support this suggestion and indicate that at 70 kgauss 87% of the electrons are in the superconducting state at 0°K. The height of the heat capacity peak in zero field relative to the heat capacity of the normal state,

$$\Delta C_p/\gamma T_c = 1.87, \tag{5}$$

is in reasonable agreement with the values of 1.52 and 3.0 given by the BCS[9] and Gorter-Casimir theories, respectively. No agreement at all is obtained with the value of 4000 calculated from the Rutgers formula[10] or with any formulation which assumes that the magnetic field is entirely excluded from the sample.

The results of the magnetic measurements are shown in Fig. 2. Each point was taken by lowering

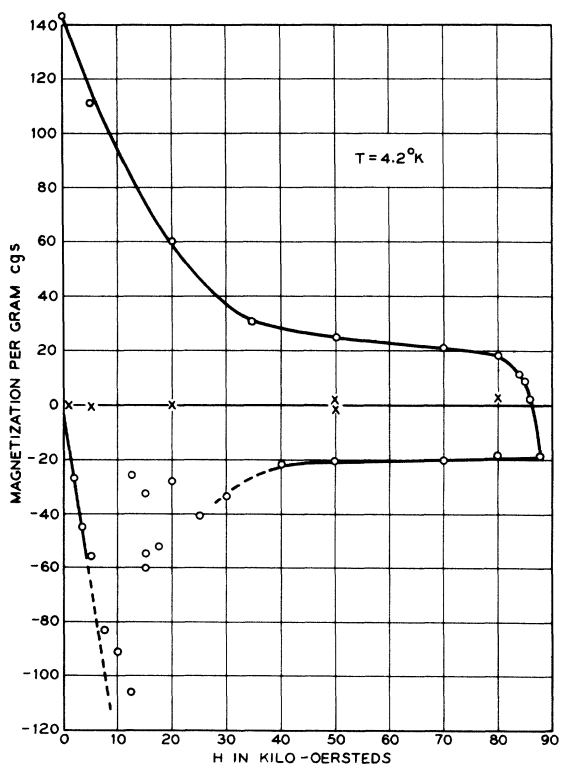

FIG. 2. The magnetization as a function of magnetic field for one-half of the sample used in the heat capacity measurements. Although the magnetization curves depend on the size of the sample, the data (open circles) show that the magnetic field is excluded from only a small fraction of the material at high fields. The crosses indicate the absence of a Meissner effect when the sample is cooled in a magnetic field.

the sample into a probe coil contained in the cryostat, cooling the sample to 4.2°K, pulling the sample out of the coil, and observing the deflection of a galvanometer connected to the coil. The open circles were taken by cooling in zero field and then bringing the field up to the desired value. Interpreted in the usual way, these experiments indicate that the sample is totally diamagnetic at fields up to about the calculated bulk critical field. An irregular penetration of the field begins in the region from 10 to 30 kgauss, and finally, an almost complete penetration of the field occurs from 30 to 80 kgauss. Bean[11] has pointed out that the filamentary model predicts that the magnetization of a superconductor in a magnetic field should be a function of sample dimension. This effect has been observed in V_3Ga.[12] Because of this size dependence, it is difficult to analyze our magnetic results quantitatively. However, we can draw some simple qualitative conclusions. The ratio of the magnetization at 70 kgauss to the value expected, if the entire volume excluded the field, is about 1/40. Another consequence of the size dependence is the fact that the minimum in magnetization is not a valid measure of the thermodynamic critical field, since the position of the minimum also depends upon sample size. When the magnetic field was decreased from the highest field point shown in Fig. 2, an increase in magnetization was obtained as expected from trapped flux in a filamentary superconductor.

The data represented by the crosses were obtained while checking for a Meissner effect by cooling the sample in a magnetic field. The absence of appreciable expulsion of flux in highly filamentary materials is not surprising. Regardless of how small the magnetic field is, the sample must pass through the filamentary state as it is cooled from above the critical temperature. Once the filamentary state is formed, and flux is trapped, an appreciable Meissner effect is impossible.

All of the results reported here can be interpreted by assuming that the sample contains a large number of filaments (probably dislocations) whose effective diameters are sufficiently large (but less than the penetration depth) that most of the sample appears superconducting. Because of the structure and low compressibility of V_3Ga, this assumption has been shown to be reasonable by Hauser and Helfand.[5] However, it is expected that a perfect single crystal of V_3Ga (free of dislocations) would behave more like a "soft" or nearly ideal superconductor and have a critical field of the order of 6 kgauss at 0°K.

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[10]See reference 2, p. 60.

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