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Bulk Upper Critical Field of Clean Type-II Superconductors: V and Nb

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The bulk upper critical field H_{c2} of single crystals of Nb and V has been measured for various crystallographic orientations within the temperature range between the respective critical temperature and $T \approx 0.06$ °K. The normalized critical field averaged over all crystallographic directions $\langle h(t)\rangle = \langle H_{c2}(t)\rangle/(-dH_{c2}/dt)_{t=1}$, where $t = T/T_c$, and the relative anisotropy of H_{c2} for the two metals are found to be similar. The measurements support predictions of the Hohenberg-Werthamer calculation that attributes the observed anisotropy of H_{c2} to the effects of an anisotropic Fermi surface. Some differences in the behavior of $H_{c2}(t)$ for the present samples can be understood qualitatively to arise from the effects of a shorter electron-collision time τ for the less pure V specimen. At very low temperatures, $H_{c2}(0) - H_{c2}(t)$ for V does not follow a $t^2\ln(t)$ temperature dependence, previously observed for Nb, but can be characterized by an empirical formula $t^2\ln(t+\alpha)$ in which the constant α is conjectured to be inversely proportional to τ.

I. INTRODUCTION

The characteristics of superconductivity in Nb and V have attracted considerable interest because these metals (and possibly La[1]) are the only known elemental superconductors which are type II for all temperatures below their respective transition temperatures. Therefore, pure samples have to date provided the largest ratio of electron mean free path to coherence length l/ξ_0 for a type-II system and are consequently important for testing the validity of "clean-limit" theories. Discovery by Tilley *et al.*[2] that H_{c2} of Nb is anisotropic demonstrated that $H_{c2}(T)$ in the clean limit may reflect "real-metal" effects not included in the Ginzburg-Landau-Abrikosov-Gor'kov (GLAG) theory which has been so successful in describing the superconducting characteristics of alloys.[3] Qualitative agreement between the data of Reed *et al.*[4] for the anisotropy of H_{c2} of pure Nb and predictions for the temperature dependence near T_c as developed in the calculations by Hohenberg and Werthamer[5] (hereafter, HW) for the effects of Fermi-surface anisotropy suggested that the previously observed discrepancy[6–13] between the critical field at low temperatures and that predicted by the Helfand-Werthamer[14] exact solution to the GLAG theory may also arise from such real-metal effects. The possible importance of Fermi-surface effects on H_{c2} of Nb was first established on a quantitative basis by Mattheiss[15] who evaluated the HW expression for the normalized and crystallographically averaged critical field at zero temperature,

$$\langle h(0)\rangle = -\langle H_{c2}(0)\rangle/H'_{c2}(1)\,,$$

where

$$H'_{c2}(1) = \left(\frac{dH_{c2}}{dt}\right)_{t=1}$$

and $t = T/T_c$. Employing an augmented-plane-wave (APW) band model for Nb, Mattheiss's calculations indicated that $\langle h(0)\rangle$ should be enhanced ~ 30% over the value given by the free-electron model. The predicted value of $\langle h(0)\rangle = 0.99$ was in good agreement with the experimental value of 0.96 ± 0.03 reported by Williamson and Valby.[16] Thus, the first quantitative test of the nonlocal theory showed that the two-band model,[17] which includes effects of coupling between s- and d-band electrons, need not be invoked to explain the observed $\langle h(0)\rangle$.

In this paper, I report the results of measurements of the temperature dependence and anisotropy of H_{c2} for single crystals of V and compare them with the behavior observed for Nb. Since V shares the same column of the periodic table as Nb and since certain aspects of the topology of the Fermi-surfaces for the two metals are reported from magnetoresistance studies[18] to be similar, it might be expected that the Fermi surfaces exhibit similar anisotropy and thus a similar behavior of the normalized superconducting properties such as $\langle h(t)\rangle$ and the relative anisotropy of $H_{c2}(t)$.

Radebaugh and Keesom[13] have reported that

$$\kappa_1 = H_{c2}(t)/\sqrt{2}\, H_c(t)$$

for H along the [110] direction of V and for temperatures as low as $T = 0.5\,^{\circ}\mathrm{K}$ is similar to the behavior reported by other researchers for Nb; and more recent magnetization measurements by Williams and Court[19] on a "relatively impure" V sample have shown a qualitative similarity with Nb for the relative anisotropy of H_{c2} within the temperature range $1.7\,^{\circ}\mathrm{K} \leq T \leq 4.2\,^{\circ}\mathrm{K}$. The present results demonstrate that the relative anisotropy of fairly pure V($\Gamma \equiv \rho_{295\,^{\circ}\mathrm{K}}/\rho_{4.2\,^{\circ}\mathrm{K}} = 140$) is comparable to that of the purest Nb investigated to date ($\Gamma = 750$ and $\Gamma = 1600$) and that $\langle h(t)\rangle$ for the two superconductors is similar. Measurements of H_{c2} in Nb and V have been extended down to temperatures as low as $0.06\,^{\circ}\mathrm{K}$. The results are consistent with the notion that Fermi-surface anisotropy is responsible for both the H_{c2} anisotropy and the enhancement of $\langle h(t)\rangle$ at low temperatures for both Nb and V. Differences in the temperature dependence of the relative anisotropy and $\langle h(t)\rangle$ between V and Nb can be qualitatively explained by the effects of a shorter electronic-collision time in the less pure sample of V.

Section II will briefly describe experimental details for the critical-field measurements. This will be followed by a comparison of $\langle h(t)\rangle$ and the relative anisotropy for $0.06\,^{\circ}\mathrm{K} \leq T \leq T_c$. The paper is concluded with a detailed discussion of the results obtained at very low temperatures.

II. EXPERIMENTAL DETAILS

This study focused on the behavior of the critical fields of two single-crystal specimens, one of V and one of Nb, each cylindrical in shape and seeded so that the $[1\bar{1}0]$ direction was within 2° of the cylinder axis. The V double-refined starting material, kindly supplied by the U. S. Bureau of Mines, was electron-beam zone refined at a pressure of 10^{-7} Torr and etched to remove 0.02 cm of the surface layer. A final heat treatment at $1500\,^{\circ}\mathrm{C}$ at a pressure of 10^{-6} Torr was applied in an attempt to reduce the dislocation density. The resulting single crystal of 0.3 cm diam and 1.5 cm length had a resistance ratio $\Gamma = 140$. Starting material for the Nb crystal was Union Carbide electrodeposited powder. After swaging, the rod-shaped compacted powder was induction melted and zone refined at a pressure of 10^{-6} Torr. This was followed by electron-beam zone refining at 6×10^{-8} Torr and a high-temperature anneal at $2120\,^{\circ}\mathrm{C}$ sustained for several days at a pressure of 10^{-9} Torr. The resulting single crystal had dimensions similar to those of the V specimen, but the resistance ratio was considerably higher, $\Gamma = 750$.

The upper critical field H_{c2} of each specimen was detected by the change in magnetic susceptibility as an external dc field was slowly swept through the region of that second-order transition. A modulation coil provided a 400-Hz field of 0.1 – Oe or less amplitude parallel to the sample's axis, and the transition was monitored by phase-sensitive detection of the resulting voltage from a pickup coil encircling the sample. Thermal-conductivity measurements of the Nb sample *in situ* at $T = 4.2\,^{\circ}\mathrm{K}$ demonstrated that the transition which was detected magnetically was a bulk transition and thus defined the bulk upper critical field H_{c2}. The width of the magnetic transition in most cases was slightly less than 1% of $H_{c2}(T)$ and therefore limited the accuracy with which the value of H_{c2} could be established; however, the relative precision of determining changes in H_{c2} with respect to changes in T or orientation was generally better than 0.1%.

A dc magnetic field sufficient to drive the sample normal was provided by a rotatable superconducting "Helmholtz"-coil pair,[20] a similar copper-coil pair, or a 60-kOe superconducting solenoid. In all cases, the field was applied in a plane perpendicular to within 2° of the axis of the sample. The ratio of field to current for these magnets had been calibrated against a 0.1% Rawson-Lush rotating-coil gaussmeter, and the field in the superconducting magnets was continously monitored during experiments by a Hall probe near the sample in order to avoid spurious effects such as might be associated with flux trapping in the superconducting magnets. A check on the accuracy of the field calibration was provided by measurements on Nb for $1.5\,^{\circ}\mathrm{K} \leq T \leq 4.2\,^{\circ}\mathrm{K}$ in an electromagnet while simultaneously monitoring the field with the 0.1% gaussmeter.

Refrigeration of the sample for measurements within the range $0.38\,^{\circ}\mathrm{K} \leq T \leq 2\,^{\circ}\mathrm{K}$ was provided by a "one-shot" ^{3}He cryostat. The specimen was bound to the ^{3}He pot by loops of 0.08-mm-diam insulated copper wire and General Electric 7031 varnish. A 10-Ω $\frac{1}{8}$-W Allen Bradley resistor mounted on the sample was employed to monitor possible heating effects from the modulation field. Thermometry was obtained from carbon resistors calibrated *in situ* against the ^{3}He vapor pressure corrected for the thermomolecular pressure ratio appropriate for a 0.3-cm-diam stainless steel tube. For $T > 0.4\,^{\circ}\mathrm{K}$, temperatures indicated by this method agreed to within 2% with the magnetic temperature of powdered cerous magnesium nitrate (CMN) packed into a form whose shape was that of a right circular cylinder of diameter equal to height.

Experiments for $0.055\,^{\circ}\mathrm{K} \leq T \leq 0.5\,^{\circ}\mathrm{K}$ were performed with a ^{3}He-^{4}He dilution refrigerator which was slightly modified from the design of Wheatley *et al.*[21] As is illustrated in Fig. 1, the bottom of the mixing chamber contained a 0.6-cm-thick layer of sintered copper to provide good

thermal contact between the helium and the copper chamber wall. One end of each of six 0.8-mm-diam copper wires was tungsten-inert-gas welded to a tab on the bottom of the copper chamber, and seventy 0.05-mm-diam copper wires were similarly attached to the other end. One-third of these thinner wires led to a copper thermal guard at the bottom of the sample mount as illustrated in Fig. 1. The guard, which served as a mount for a teflon "spider" used to center the sample within the thermal shield, ensured that a minimal temperature gradient would exist between the sample and mixing chamber. The remaining two-thirds of the 0.05-mm-diam wires passed through the CMN thermometer, a slurry of powdered CMN and Apiezon "*J*" oil, and down to the sample mount, a copper clamp. One end of the sample, when clamped and subsequently varnished, was thereby thermally tied to the mixing chamber. To the other end was clamped a 200-Ω $\frac{1}{4}$-W Speer-type 1002 resistor whose value, compared with that of a similar resistor mounted inside the mixing chamber, indicated the sample's temperature and the magnitude of thermal gradients. The resistors were calibrated *in situ* with the Helmholtz-coil pair removed. For present purposes, the uncertainty in determining temperature introduced by the small magnetoresistance of the resistors was negligible.

Thermometry in the high-temperature region above 4.2 °K was based upon the manufacturer's calibration of a germanium resistance thermometer.[22] Carbon resistors were used as secondary thermometers when the applied magnetic field was sufficiently high to cause an appreciable magnetoresistance in the germanium. Calibration of the resistance thermometer was checked by measuring the critical temperature of a pure sample of Pb. The observed value of $T_c = (7.20 \pm 0.01)$ °K compares favorably with $T_c = 7.195$ °K reported by Finnemore *et al.*[8] and 7.193 °K by Franck and Martin.[23] For the Nb sample, $T_c = (9.27 \pm 0.02)$ °K, in agreement with $T_c = (9.25 \pm 0.01)$ °K of Finnemore *et al.*[8] and $T_c = 9.26$ °K by Shen *et al.*[24] For V, $T_c = (5.385 \pm 0.015)$ °K is also in accord with $T_c = 5.414$ °K reported by Radebaugh and Keesom.[13]

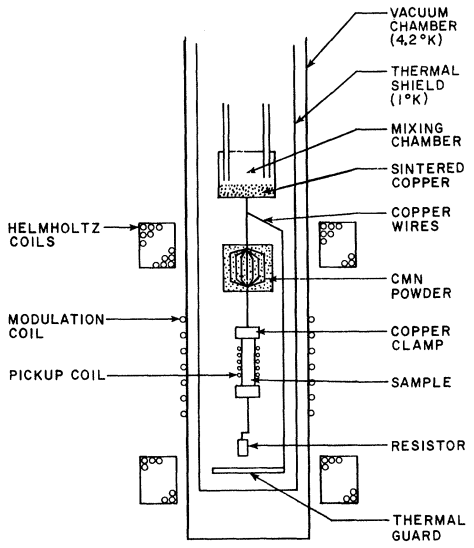

FIG. 1. Schematic of the very-low-temperature portion of the ^{3}He-^{4}He dilution refrigerator showing the thermal linkage between sample and mixing chamber.

III. ANISOTROPY

Since both Nb and V are bcc crystals, the observed orientation dependence of H_{c2} can be described in a natural way by a series of "cubic harmonics," an orthogonal set of functions which are the identity representation of the full cubic group O_h,

$$H_{c2}(t, \theta, \phi) = \sum_{n=1}^{N} A_n(t) K_n(\theta, \phi) \ . \tag{1}$$

Several cubic harmonics $K_n(\theta, \phi)$, expressed not in terms of the spherical coordinates θ and ϕ but in terms of the direction cosines with respect to the principal axes, have been tabulated by Von der Lage and Bethe.[25] As employed in Eq. (1), these functions are normalized to a value of 4π when their squared modulus is integrated over all θ and ϕ. More complete listings of these functions have been given by Altmann and Cracknell,[26] Altmann and Bradley,[27] and Mueller and Priestley.[28] Each cubic harmonic can be represented by an expansion in a series of spherical harmonics Y_l^m of a given l; the values of l appropriate to our notation of Eq. (1) for n running from $n = 1$ through $n = 5$ are $l = 0, 4, 6, 8, 10$. There is no ambiguity in this notation for $l \leq 10$; however, for $l > 10$, two or more values of n may be associated with different spherical harmonic expansions of a given l.

The anisotropy of H_{c2} was measured with H in the $(1\bar{1}0)$ plane of the samples. Parameters A_n of Eq. (1) were then adjusted by computer so as to obtain a least-squares fit to the data at each temperature. Presumably, Eq. (1) represents a converging series, so that the consequence of employing a finite number of terms can be judged from the results of computer fits for successively larger N. For V, $N = 4$ sufficed for an adequate description at all temperatures as, for example, is shown in Fig. 2 for $t = 0.072$. However, for Nb, systematic deviations from the $N = 4$ fit could be observed for $T \lesssim 2.8$ °K as was first reported by Farrell *et al.*[29] for a less pure Nb sample of $\Gamma = 100$. An example of the quality of the $N = 4$ fit is illustrated in Fig. 3 for $t = 0.04$. It was found that only slight improvement resulted from a fit for $N = 5$. In view

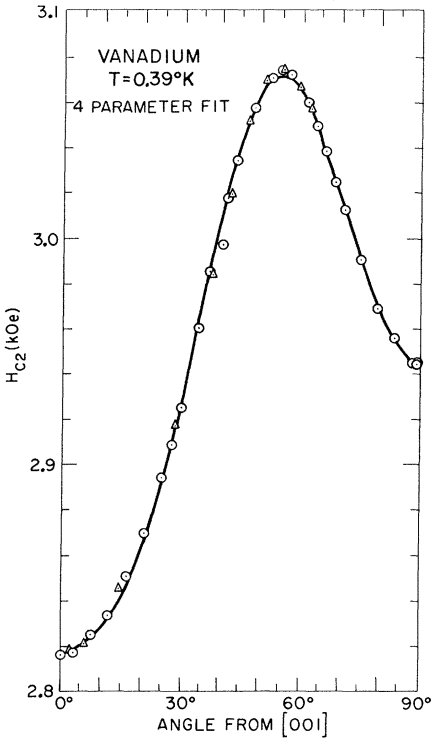

FIG. 2. Orientation dependence of H_{c2} in the $(1\bar{1}0)$ plane of V for $t = 0.072$. The solid line represents a fit by four cubic harmonics as explained in the text.

of the difficulty of establishing the value of A_5 with high confidence because of a possible 0.1% systematic error arising from sample misalignment, and since values of the higher-order harmonic coefficients seem to be sensitive to the electron-collision time,[29] it was judged that the present experimental precision and sample quality did not warrant a five-term expansion for Nb.

A. Average Critical Field

The critical field when averaged over all crystallographic directions is given by

$$\langle H_{c2}(t)\rangle = A_1(t) \tag{2}$$

since $K_1(\theta, \phi) = 1$. Perhaps a more useful quantity for comparing critical fields of different superconductors is the reduced field

$$\langle h(t)\rangle = -A_1(t)/H'_{c2}(1)$$

introduced by Helfand and Werthamer[14] and generalized by HW. For isotropic superconductors, $h(t)$ was predicted[14] to be a universal curve which is sensitive only to the electronic-collision time, providing strong-coupling and paramagnetic-limiting effects are absent. The $\langle H_{c2}(t)\rangle$ data for Nb and V are therefore normalized to the present empirical values

$H'_{c2}(1) = -(4.42 \pm 0.08)$ kOe and $-(3.28 \pm 0.06)$ kOe, respectively, the latter being about 6% lower than the value reported by Radebaugh and Keesom[13] for data with H along the [110] direction. Only $H_{c2}(t)$ data for $0.92 \leq t \leq 1$ were utilized to determine the slopes, a region where the anisotropy for Nb was less than 1.5% of $H_{c2}(t)$ and where the critical field for the [001] direction varied linearly with t. Table I summarizes pertinent characteristics of the Nb and V samples.

Figure 4 shows the present results for $\langle h(t)\rangle$. The curves for Nb and V both lie some 30% above the Helfand-Werthamer clean-limit prediction for an isotropic superconductor at low temperatures. It is remarkable that for a wide temperature range the observed curves lie *above* the straight line extrapolated from the slope near $t = 1$. Such behavior for $H_{c2}(t)$ along [110] of V is indicated by Radebaugh and Keesom's observation[13] that their data for $t \approx 1$ can best be described by a formula which exhibits positive curvature. This positive curvature is qualitatively different from the ideal behavior of bulk *dirty* type-II superconductors, which in the absence of paramagnetic effects have critical field curves that at all temperatures show nonpositive curvature.

The value of $\langle h(t)\rangle$ for V differs from that of Nb

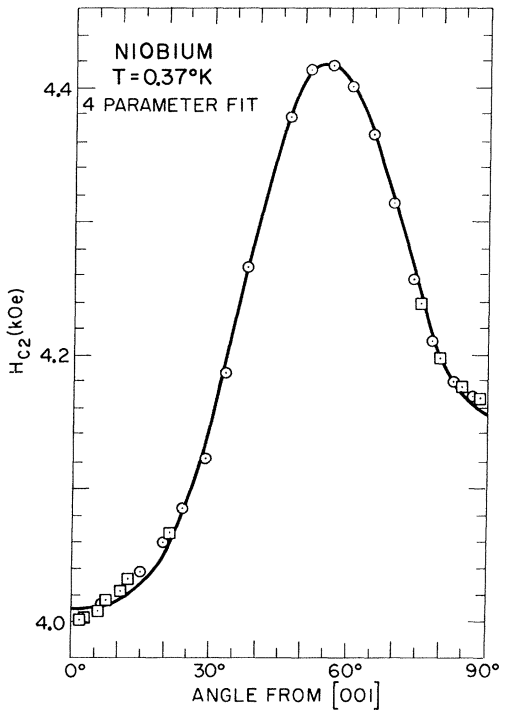

FIG. 3. Orientation dependence of H_{c2} in the $(1\bar{1}0)$ plane of Nb for $t = 0.05$. The solid line represents a fit by four cubic harmonics.

TABLE I. Superconductivity and electronic-structure parameters for Nb and V. Listed below are the critical temperature T_c, Debye temperature θ_D, strong-coupling parameter λ, extrapolated value of the average upper critical field at zero temperature $\langle H_{c2}(0)\rangle$, slope of the critical field $H'_{c2}(1) = (dH_{c2}/dt)_{t=1}$, normalized extrapolated field $\langle h(0)\rangle = -\langle H_{c2}(0)\rangle / H'_{c2}(1)$, and Fermi speed v.

	Nb	V	Reference
T_c	(9.27±0.02) °K	(5.385±0.015) °K	a
θ_D	277 °K	399 °K	b
λ	0.82	0.60	c
$\langle H_{c2}(0)\rangle$	(4.25±0.04) kOe	(3.02±0.03) kOe	a
$H'_{c2}(1)$	−(4.42±0.08) kOe	−(3.28±0.06) kOe	a
$\langle h(0)\rangle$	0.96±0.03	0.92±0.03	a
$\langle v^2\rangle^{1/2}$	2.94×10^7 cm/sec	1.80×10^7 cm/sec	d
$\langle v^2\rangle^{1/2}$	3.45×10^7 cm/sec	···	e

[a] Present results.

[b] F. Heininger, E. Bucher, and J. Müller, Physik Kondensierten Materie 5, 243 (1966).

[c] Reference 30.

[d] Obtained from $H'_{c2}(1)$ and Eq. (5).

[e] Reference 15 for which the APW band-model Fermi speed $\langle v_B^2\rangle^{1/2}$ has been reduced by the phonon renormalization factor $(1+\lambda) = 1.8$ for Nb.

by less than 4% over the entire temperature range. Because of a ±1% uncertainty in measuring $H_{c2}(T)$ and a ±2% uncertainty in $H'_{c2}(1)$, it cannot be stated with absolute certainty that the difference between the two $\langle h(t)\rangle$ curves is significant; however, it would seem unlikely that experimental errors are solely responsible for the 4% difference. Because the V sample has a much smaller Γ than does the Nb, a shorter electron-collision time for V could qualitatively explain why the V curve lies closer to the dirty-limit prediction.

The HW calculation for the enhancement of $\langle h(0)\rangle$ due to the effects of Fermi-surface anisotropy predicts that in the limit of small anisotropy for H_{c2},

$$\langle h(0)\rangle = 0.7273 \exp\left[-\int d\hat{q}\, \frac{N(\hat{q})}{\langle N\rangle} \ln\left(\frac{v^2(\hat{q})}{\langle v^2\rangle}\right)\right] \quad , \quad (3)$$

where $N(\hat{q})$ is the density of electronic states at the Fermi surface for the direction $\hat{q}$ in momentum space and $v(\hat{q})$ is the Fermi speed for a given $\hat{q}$. Mattheiss[15] has evaluated the integral for an APW band model for Nb and found $\langle h(0)\rangle = 0.99$, a value which Williamson and Valby[16] showed to be in accord with the behavior of the present Nb sample, $\langle h(0)\rangle = 0.96 \pm 0.03$. The close agreement is remarkable in view of the neglect in Eq. (3) of the effects of anisotropic electron-phonon coupling. It might be suspected that such an anisotropy is important since the zero-field gap is reported to have a ~10% anisotropy, [30] but whether this theoretically should appreciably affect the *average* upper critical field is not clear. Agreement between experiment and Mattheiss's results indicates that it does not. Close agreement between $\langle h(0)\rangle = 0.92 \pm 0.03$ for V and the Nb result supports the hypothesis that the relative anisotropy of the Fermi surfaces, as defined by the integral in Eq. (3), is similar, even though the average Fermi speed for the two metals differs substantially (Table I).

Since T_c/θ_D for V is 2.5 times the value for Nb, strong-coupling effects between electrons and phonons should substantially differ, as is indicated by the values of the strong-coupling parameter λ derived by McMillan[31] (see Table I). Similarity between the Nb and V curves of Fig. 4 supports the theoretical conclusion of Werthamer and McMillan[32] that strong-coupling effects should play only a minor role in determining $h(t)$.

Another quantitative prediction of the HW calculation and Mattheiss's band model can be compared with our data for Nb and V; in the weak-coupling limit, [5]

$$H'_{c2}(1) = -\frac{6c(2\pi k T_c)^2}{7\hbar e \zeta(3)\langle v^2\rangle} \quad , \quad (4)$$

where e is the magnitude of the electron charge and c is the speed of light. For this equation it must be remembered that v is the actual Fermi speed, assumed to be related to a band-model Fermi speed v_B by $v = v_B(1+\lambda)^{-1}$. Eilenberger and Ambegoakar[33] have shown that Eq. (4) should be modified for strong-coupling superconductors to include the term in parentheses:

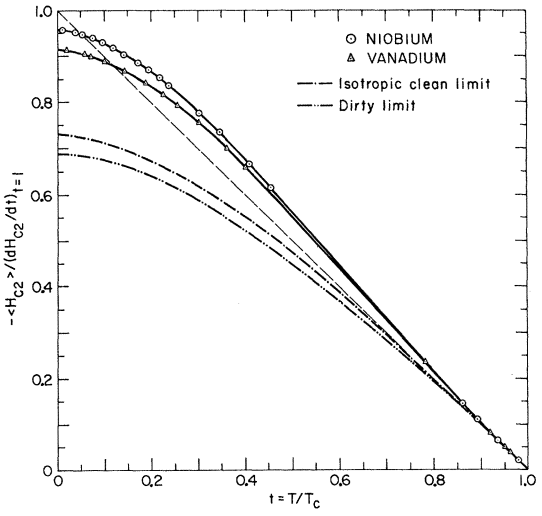

FIG. 4. Observed critical field for Nb and V averaged over all crystallographic directions and normalized to the slope at $t \approx 1$. Predictions of Ref. 14 for the behavior in the clean and dirty limit for an isotropic Fermi surface are also shown, as is the linear extrapolation of the behavior at $t \approx 1$.

$$H'_{c2}(1) = -\frac{3.15\times10^{16}T_c^2}{\langle v^2\rangle}\left(2\frac{\Delta^2_{\rm OBS}}{\Delta^2_{\rm BCS}} - \frac{H^2_{c,\rm OBS}}{H^2_{c,\rm BCS}}\frac{\Delta^2_{\rm BCS}}{\Delta^2_{\rm OBS}}\right)_{t\to1}. \quad (5)$$

The first numerical factor gives the value of the constants of Eq. (4). The strong-coupling factor in parentheses contains the ratios of observed to BCS-model energy gap Δ and thermodynamic critical field H_c. McMillan and Hohenberg[34] have estimated that the value of this factor for Nb is 1.4; whereas for V, I estimate from the known ratio of T_c/θ_D that it is 1.16. Clearly, from a theoretical standpoint, strong-coupling effects can greatly affect the *magnitude* of $H_{c2}(t)$, even though the present experimental results indicate that the *normalized* field $\langle h(t)\rangle$ is not substantially affected.

Mattheiss's band model for Nb predicts, according to Eq. (5), that $H'_{c2}(1) = -3.14$ kOe. This value is ~ 30% below the experimental result

$$H'_{c2}(1) = -(4.42\pm0.08)\ \text{kOe},$$

but better agreement could be obtained by a reduction in the theoretical $\langle v_B^2\rangle$ which would result from a decrease in the width of the d band. Mattheiss[15] has indicated that such a change in the model would not appreciably affect $\langle h(0)\rangle$ and would have the additional advantage of improving agreement between the band density of states at the Fermi level and the density of states indicated by specific-heat measurements. In view of the difficulty of obtaining an accurate value of $\langle v^2\rangle$ from transport and magnetic measurements, it may be useful to accept the applicability of Eq. (5) and thereby find from the measured value of $H'_{c2}(1)$ that

$$\langle v^2\rangle^{1/2} = 2.94\times10^7\ \text{cm/sec}.$$

For V, no significant test of Eq. (5) is possible at present, although the measured slope

$$H'_{c2}(1) = -(3.28\pm0.06)\ \text{kOe}$$

would imply

$$\langle v^2\rangle^{1/2} = 1.80\times10^7\ \text{cm/sec},$$

a value identical to that obtained by Radebaugh and Keesom[13] from transport and superconductivity parameters. The large difference in Fermi speed for Nb and V is evidence that substantial differences do exist between the band structures near the Fermi surfaces of the two metals and that therefore the band model and Fermi level for one cannot be taken to be representative of the other. In view of the present uncertainties in the Nb band model, the discrepancy between the experimental and theoretical value for $H'_{c2}(1)$ is presently not regarded as serious.

B. Higher-Order Coefficients

Experimental results described in Sec. III A have shown that normalized quantities such as $\langle h(t)\rangle$ are similar for Nb and V, whereas unnormalized quantities such as $H_{c2}(t)$ are not. In the comparative study of the anisotropy coefficients of Nb and V it was therefore decided to plot the normalized coefficients $A_2(t)/\langle H_{c2}(t)\rangle$ and $A_3(t)/\langle H_{c2}(t)\rangle$ which would provide a measure of the *relative* anisotropy. These parameters are shown in Figs. 5 and 6 for representative temperatures. Also shown for comparison are the published results of Reed *et al.*[4] for a purer sample of Nb($\Gamma = 1600$) and points taken from smooth-curve fits to the results of Farrell *et al.*[29] for a less pure sample of Nb($\Gamma = 100$). Reed *et al.*[4] employed a three-parameter fit to data obtained with H along the [111], [110], and [001] crystallographic directions, whereas Farrell *et al.*[29] utilized a least-mean-squares fit to data obtained with H in the $(1\bar{1}0)$ plane. Most of the indicated "three-parameter" data points from the present study were obtained by the same procedure as used by Reed *et al.*; however, when a "four-parameter" point is also shown for a given temperature, both points were obtained from a least-squares fit to data for the $(1\bar{1}0)$ plane. It should be noted that the present choice of normalization for the cubic harmonics yields values of A_2 and A_3 that differ from those of Refs. 4 and 29 by multiplicative constants, and that data from these two references have been appropriately renormalized for inclusion in Figs. 5 and 6.

Perhaps the most striking feature of the data in Fig. 5 is the similarity in the behavior of $A_2/\langle H_{c2}\rangle$ for Nb($\Gamma = 750$), Nb($\Gamma = 1600$), and V($\Gamma = 140$). Agreement between the higher-purity Nb samples – and by implication insensitivity to the electron-collision time – suggests that the Nb sample of the present study does in this sense represent the clean limit. Thus, the smaller anisotropy for Nb($\Gamma = 100$) is consistent with the observation by Reed *et al.*[4] that H_{c2} anisotropy is reduced by a shorter electron-collision time. Similarly, it is reasonable to expect that V($\Gamma = 150$) would have an anisotropy also reduced by the relatively short collision time. Agreement between the V($\Gamma = 150$) and pure Nb data in Fig. 5 is therefore regarded as fortuitous; consequently, it is likely that purer V will display a larger relative anisotropy than pure Nb.

No calculation is known that predicts the value of $A_2/\langle H_{c2}\rangle$ for all temperatures in terms of material parameters. The solid line proportional to $(1-t)$ in Fig. 5 is an extension of the high-temperature linear trend predicted by Hohenberg and Werthamer[4] for $t \approx 1$. To a good approximation, this trend is obeyed in Nb down to the lowest temperature employed, although as yet there is no theoretical justification for this behavior. An indication of the uncertainty in the data can be obtained both from the scatter of the points and from the change which results from a

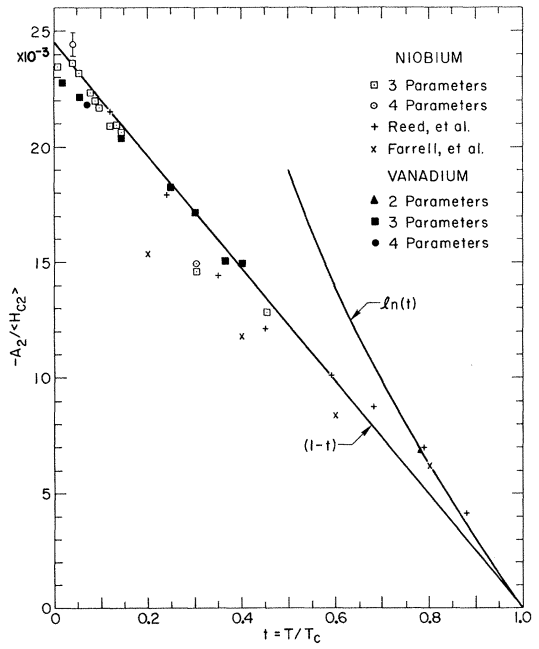

FIG. 5. Temperature dependence of the coefficient A_2 of Eq. (1) normalized to the average critical field. The present results are compared with those of Reed *et al.* (Ref. 4) for a purer sample of Nb ($\Gamma = 1600$) and with those of Farrell *et al.* (Ref. 29) for a less pure Nb sample ($\Gamma = 100$).

four-parameter versus three-parameter fit, as is shown for several temperatures. Takanaka and Nagashima[35] have emphasized that $A_2/\langle H_{c2}\rangle$ theoretically should not have a linear dependence on t for temperatures much below T_c; however, their alternate proposal of a $\ln(t)$ dependence is shown in Fig. 5 to be not as successful over a wide temperature range as the linear approximation.

Data for $A_3/\langle H_{c2}\rangle$ are displayed in Fig. 6. The trends for various samples are consistent with the conclusion of Farrell *et al.*[29] that coefficients of higher-order cubic harmonics are more sensitive to decrease in τ than lower-order coefficients: (i) V($\Gamma = 140$) data now fall below the purer Nb data and (ii) Nb($\Gamma = 100$) data are substantially below the purer Nb data. The fact that the V($\Gamma = 140$) data now agree with the Nb ($\Gamma = 100$) behavior is apparently another coincidence.

No detailed study of the temperature dependence of A_4 has been made because the V sample appears not to represent the clean limit, and more than five harmonics are required to describe adequately the Nb behavior at low temperatures. Takanaka and Nagashima[35] have predicted that A_3/A_4 ought to be independent of t for $t \approx 1$. The results of Farrell *et al.*[29] are in approximate agreement with this prediction, but the temperature insensitivity of the ratio was also observed to extend to relatively low temperatures. For V, I find $A_3/A_4 = 2.7$ at $t = 0.07$ and for Nb, $A_3/A_4 = 3.7$ at $t = 0.04$. To what extent this ratio depends upon Fermi-surface parameters and to electron-collision time is presently unknown; however, the fact that A_2/A_3 is larger for V than Nb whereas A_3/A_4 is smaller implies that the ratios of successive coefficients are not determined solely by a monotonically decreasing function of only the collision time.

IV. VERY-LOW-TEMPERATURE RESULTS

As H_{c2} at low temperatures becomes relatively insensitive to temperature, special attention was devoted toward maximizing the relative precision of the critical-field measurements so that meaningful conclusions about the limiting temperature dependence could be drawn. Thus, although the finite width of the observed magnetic transition gave an uncertainty of about 1% to the accuracy of establishing H_{c2}, if an arbitrarily selected feature of the transition were consistently used to define H_{c2}, the precision in determining the field for this feature was generally ~ 0.1% during a given experimental run. Comparison of the data taken with the ^{3}He cryostat and that obtained with the same specimen remounted in the ^{3}He-^{4}He dilution refrigerator could show as much as a 0.5% discrepancy in H_{c2} along a principal crystallographic direction in the region of common sample temperature. This discrepancy was attributed to a change in the trap-

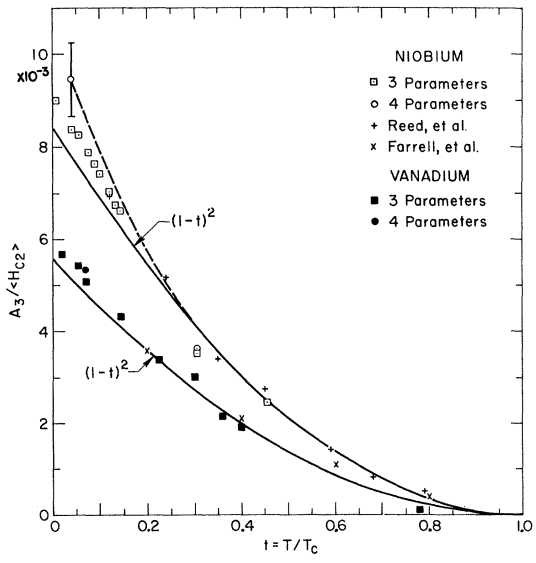

FIG. 6. Temperature dependence of A_3 normalized to the average critical field. For Nb, a three-parameter fit results in a ~10% error for A_3 at the lowest temperatures compared with a four-parameter fit. For V, the error is considerably less. The present data are compared with those of Refs. 4 and 29.

ped flux during a thermal recycling of the superconducting magnet and was eliminated by an additive term in H_{c2} to match the very-low-temperature results to that observed at higher temperatures. In addition, a multiplicative ~0.1% correction may be required for the low-temperature values of H_{c2} for the two remaining crystallographic directions in order to compensate for slight misorientation of the sample. As these adjustments were employed only to match one experimental run to another and were temperature independent, they cannot affect conclusions drawn about the temperature dependence of H_{c2}.

Figures 7 and 8 illustrate the very-low-temperature data for Nb and V, where data indicated by triangles were obtained by use of the ^{3}He-^{4}He dilution refrigerator. In contrast to a quadratic temperature dependence for $H_{c2}(0)$-$H_{c2}(t)$ as previously observed at low values of t for Zr-Nb dirty alloys,[36] Fig. 7 shows that these clean specimens of Nb and V can be described by a quadratic dependence only for a very limited range in t^2, as is indicated by the solid lines. The less pure specimen (V) of Fig. 7 has a critical field which is consistent with a quadratic dependence up to higher values of t than is the purer specimen (Nb).

Williamson and Valby[16] have recently reported that H_{c2} of the present Nb specimen is accurately described for $t<0.17$ by a dependence of the form

$$H_{c2}(t)/H_{c2}(0)=1+\eta t^2\ln(t)\ , \tag{6}$$

where η and $H_{c2}(0)$ depend upon crystallographic direction. This is illustrated in Fig. 8, where the abscissa includes approximately the same range in t as is shown in Fig. 7. However, Fig. 8 also reveals that the dependence of the form of Eq. (6) is inadequate over the same range in t for the present V data.

Gor'kov[37] has predicted that Eq. (6) with $\eta=0.65$ should be valid for isotropic clean type-II superconductors in an approximation that neglects terms of $O(t^2)$. Although Gor'kov employed a variational method to calculate the critical field, Helfand and Werthamer[14] have shown that his gap-nucleation function is, in fact, the correct solution. As is discussed in Ref. 16, the logarithmic factor is based upon general considerations of the nonlocality of superconductivity in pure metals and therefore might be expected to obtain even in anisotropic superconductors such as Nb and V. Equation (6) implies that the free energy at H_{c2} is nonanalytic at $T=0$, a feature of the clean-limit theory recently discussed by Brandt[38] in a description of the field dependence of the magnetization. However, a proper theory for real metals containing the effects of a finite collision time may remove this nonanalyticity and might account, therefore, for the deviation of the V data from Eq. (6). Maki and Tsuzuki[39] have suggested that the magnetization parameter κ_2 would not in fact diverge as their predicted clean-limit dependence $\kappa_2\sim[\ln(t)]^{1/2}$, but would be limited to

$$\kappa_2\sim[\ln(\hbar/k\tau T_c)]^{1/2}\ \text{for}\ t\to 0\ ,$$

where τ is the electron-collision time. We therefore introduce the conjecture that Eq. (6) should be generalized to include the collision time:

$$H_{c2}(t)/H_{c2}(0)=1+\eta t^2\ln(t+\hbar/k\tau T_c)\ , \tag{7}$$

where it is implicit in this equation that $H_{c2}(0)$ and η may depend upon τ. Eq. (7), which at present must be regarded as only phenomenological, removes the nonanalyticity from the theory at $t=0$ and predicts that H_{c2} will ultimately approach its zero-temperature value quadratically in t, a feature that has intuitive appeal. In the approximation Eq. (7), we neglect the temperature dependence of τ, since this equation applies only in the low-temperature realm where electron-impurity scattering is predominant.

Unfortunately, it is difficult to provide a rigorous test for Eq. (7) because of the uncertainty in evaluating the appropriate value for τ. For Nb, the Fermi speed $v\approx 2.94\times10^7$ cm/sec (Table I) agrees fairly well with the value obtained from magnetization and transport measurements[11]; however, the electron mean free path indicated by ultrasonic attenuation experiments[40] $l_u\approx 7\times10^{-4}$ cm for $\Gamma=750$ is considerably in excess of the estimate from these same magnetization and transport experiments $l_t\approx 2\times10^{-4}$ cm.[41] Taking l_u as more reliable, I estimate $\hbar/k\tau=\hbar v/kl_u\approx 0.6\,^\circ$K. Such a factor in Eq. (7) would cause a slight (0.1%) lowering of the critical-field curve in Fig. 8 for $T\lesssim 0.3\,^\circ$K $[t^2\ln(t)\lesssim 0.006]$ relative to the trend at higher temperatures and would slightly improve agreement for the Nb data as compared with a straight-line fit (solid curve) according to Eq. (6). However, the precision of the data is not sufficient to determine uniquely whether Eq. (6) or Eq. (7) is more appropriate. For Eq. (6), I find $\eta=1.69$, 1.39, and 1.30 for the [111], [110], and [001] crystallographic directions, respectively, but if Eq. (7) is applied with $\hbar/k\tau\approx 0.6\,^\circ$K, the values of η must be increased by about 20%.

For V, Fig. 8 clearly shows that Eq. (6) does not describe the data over the same range in t for which it is successful for Nb. Unfortunately, ultrasonic attenuation measurements in V have not yet been carried out, so the value of l_u is unknown. Therefore, the electron mean free path was estimated from l_u of Nb by applying a reduction factor equal to the ratio of the values of Γ for the samples, giving $l\approx 1.4\times10^{-4}$ cm. The Fermi speed obtained from $H_{c2}'(1)$ (Table I) thus yields $\hbar/k\tau\approx 1\,^\circ$K. In order to avoid a mere curve-fitting exercise for each crystallographic direction shown in Fig. 8

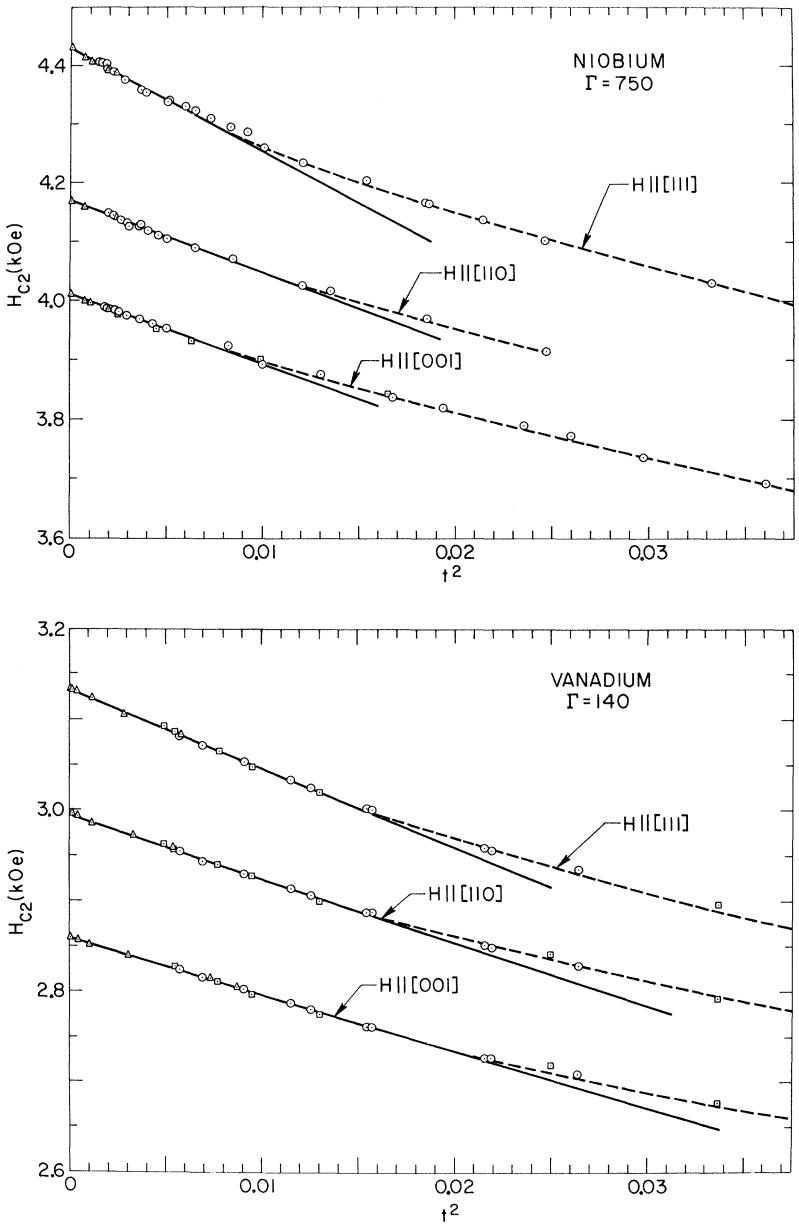

FIG. 7. Observed critical fields versus t^2 along the three principal crystallographic directions of Nb and V for temperatures up to 1.8 and 1.05 °K respectively. A quadratic approximation for the temperature dependence is successful only for $t \lesssim 0.10$ for Nb and $t \lesssim 0.13$ for V.

for V and to make the test of Eq. (7) more convincing, the number of adjustable parameters used to fit the temperature dependence of *all three* curves was reduced to one by assuming that the anisotropy of the η values for the three directions in V is equal to the anisotropy of the values for Nb. This approximation is justified solely on the basis of the close similarity discussed earlier of the H_{c2} relative anisotropy of the two metals. It is important to bear in mind that this scaling argument is only an estimate, since Figs. 2 and 3 show that the anisotropy in Nb and V is not identical. It should also be borne in mind that Gor'kov's derivation[37] of Eq. (6) for the isotropic model neglected terms of $O(t^2)$. The coefficient of the neglected t^2 term is at present unknown, so that it is not yet clear for that model – nor indeed for real metals such as Nb and V – where Eqs. (6) and (7) should cease to be accurate at high temperatures. Lacking better guidance, the η scaling parameter was determined by the criterion that the fit for V should extend over the same range in t as Eq. (6) was successful for Nb. A scaling parameter which yields the three solid curves for V in Fig. 8 is $\epsilon = 1.42$, that is, for each direction in V, $\eta^{(V)} = \epsilon \eta^{(Nb)}$. The result in Fig. 8 is a reasonably good fit to the data, and to that extent Eq. (7) is a successful phenomenological formula.

The fact that η observed for Nb and V exceeds the value predicted by Gor'kov[37] for the isotropic model indicates that this parameter may also be

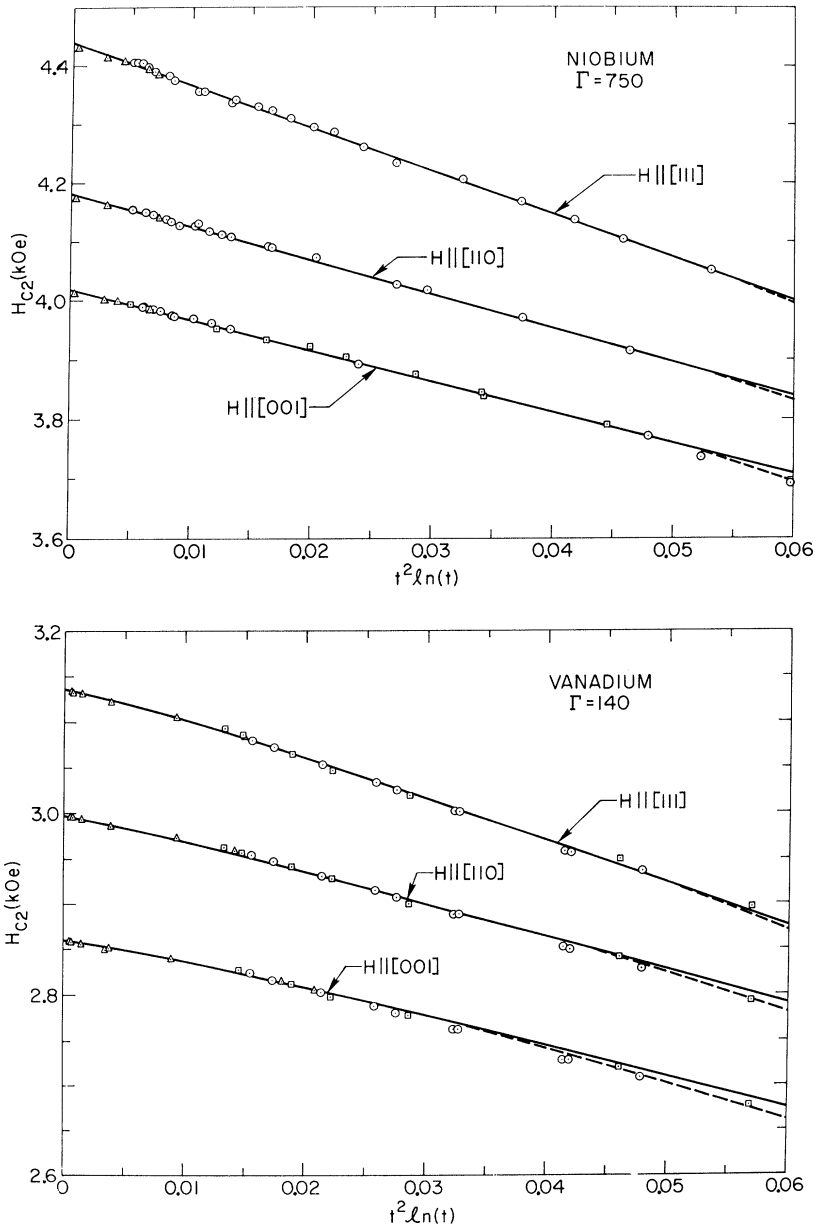

FIG. 8. Observed critical fields versus $t^2 \ln(t)$ for Nb and V for approximately the same temperature range as is included in Fig. 7. A linear relation describes the Nb behavior whereas it is inadequate for V.

affected by Fermi-surface anisotropy. Presumably η for each crystallographic direction would be calculable in terms of Fermi-surface parameters from a clean-limit theory which is valid at low temperatures and which recognizes the nonanalytic feature of the free energy at H_{c2} for $T=0$. Although η near each direction is a linear combination of the coefficients A_n, it may be that η is more readily calculable for low temperatures than the set $\{A_n(t)\}$. The success of Eq. (6) in describing thelow-temperature behavior of H_{c2} in Nb along various crystallographic directions must be reconciled with the temperature dependence of $\{A_n(t)\}$ in Eq. (1). Comments about how the logarithmic factor enters the expansion – whether in $A_1 = \langle H_{c2}(t) \rangle$ or in the remaining terms – would be only speculative at this time and should await further theoretical development. However, the present data for Nb indicate that a linear approximation for the temperature dependence of the difference between H_{c2} along [111] and [001] in Nb, which is quite successful for $T > 0.4\,^{\circ}\mathrm{K}$, [42] fails at lower temperatures. It is remarkable that η for V as determined by Eq. (7) exceeds both the free-electron value[37] and the value for Nb. The simplest explanation for this would relate a monotonic increase in η with an increase in $\langle h(0) \rangle$ due to Fermi-surface effects and suggests that $\langle h(0) \rangle$ for ideally pure V may exceed the value

for Nb.

The observed H_{c2} for Nb and V at low temperatures exhibits no pronounced change in temperature dependence in the temperature region where pure Nb samples are reported to display a change in behavior of the zero-field specific heat[24] and thermal conductivity.[43] Thus, effects of a possible small energy gap in Nb at zero field[44] do not dramatically affect the high-field behavior at very low temperatures.

V. SUMMARY

The crystallographically averaged critical field $\langle H_{c2} \rangle$ of pure Nb and V when expressed as the normalized quantity $\langle h(t) \rangle$ has been found to differ only slightly for the two metals. The anisotropy of H_{c2} when normalized to $\langle H_{c2}(t) \rangle$ is also similar. Thus, despite substantial differences in T_c, $H_{c2}(0)$, λ, and $\langle v^2 \rangle$ for the two metals and despite pronounced deviations in the behavior of the critical field with respect to the theoretical behavior of an isotropic model, the quantitative success of the Hohenberg-Werthamer calculation as evaluated by Mattheiss for Nb is compelling evidence that Fermi-surface anisotropy is responsible for the anomalous enhancement of κ_1 observed in the numerous previous experimental studies of both Nb and V. Differences in the behavior of $H_{c2}(t)$ for Nb and V can be understood qualitatively on the basis of a shorter electronic-collision time in the less pure V sample. Such effects are particularly pronounced in the very-low-temperature limit where a phenomenological formula that removes the nonanalyticity from the clean-limit theory is shown to be successful for V. Comparison of the relative anisotropy of the present Nb and V critical fields suggests that ideally pure V would have an even larger relative anisotropy than ideally pure Nb.

Note added in proof. Takanaka[45] has generalized Gor'kov's theory[37] for a clean superconductor by including the effects of an anisotropic Fermi surface. His results indicate that Eq. (6) should be valid at low temperatures. The angular variation of η with $H_{c2}(0)$ is found to be qualitatively in accord with the present experimental results.

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Magnetic Scattering of X Rays from Electrons in Molecules and Solids

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The scattering of moderately high-energy x rays from electrons in magnetic solids is analyzed. We show that (a) the incoherent Compton scattering of polarized x rays can be used to determine the spin-dependent momentum distribution function of electrons in ferromagnetic materials, and (b) the coherent Bragg scattering of unpolarized x rays can be used to determine the magnetic structure of antiferromagnetic solids below their transition temperature.

Recently[1-3] there has been renewed interest in utilizing x rays to probe the electronic properties of molecules and solids.

In the extreme nonrelativistic limit, the x rays couple exclusively to the charge of the electrons. This implies that for electrons in solids the scattering cross section is independent of the magnetic properties of the medium. However, it is well known that the complete relativistic Compton amplitude does depend on the spin of the electron.[4]

The dominant charge scattering mechanism can be thought of as arising from the acceleration of the electron by the *electric* field of the wave and the *electric dipole* reradiation of the scattered field. Although it is not quantitatively correct it is qualitatively correct to think of the spin dependence of the scattering amplitude as, at least in part, arising from the same acceleration followed by a *magnetic* dipole reradiation of the scattered field. If one thinks of the electron as a little spinning ball with a radius of the order of the Compton wavelength $\lambda = \hbar/mc$ then the ratio of magnetic dipole to electric dipole radiation is roughly $k_1 \lambda_c$, i.e., $\hbar\omega_1/mc^2$.

In this paper we will show that mildly relativistic x rays can be used to: (a) measure independently the momentum distributions of spin "up" and spin "down" electrons in magnetic solids, and (b) determine the magnetic crystal structure of antiferromagnetic solids below their transition temperatures. This magnetic Bragg scattering is analogous to conventional magnetic neutron scattering.

Since the binding energy of the outer electrons in atomic systems is small relative to typical x-ray energies, all of the physics we will discuss is contained in the formula for the scattering of light from free particles.[4] Binding effects will be in-