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Parallel critical magnetic fields for superconducting vanadium and technetium films

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The dependence of a parallel critical magnetic field $H_{c\parallel}$ for vanadium and technetium films (including ultrathin films with $d < 100$ Å) on the thickness d is investigated experimentally. The results are explained by the effect of spin phenomena on the magnitude of $H_{c\parallel}$.

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1. INTRODUCTION

The problem of the magnitude and nature of the limiting magnetic fields which restrict the existence of the superconducting state has been intensively studied, both theoretically and experimentally, in recent years, in application to both bulk[1] and film samples (see, for example, Refs. 2–5). Thin films in a parallel field are convenient objects for the study of this question for the following reasons.

The disruption of the superconducting state by a magnetic field can take place as a result of the action of the magnetic field both on the orbital and on the spin parts of the motion of the electrons. Observation of the effect phenomena on the critical magnetic field is possible in this case if H_c^{orb} (the critical field, defined under the assumption of the existence of the orbital mechanism only) has a sufficiently large value, namely, under the condition

$$\mu_B H_c^{orb}(T) \geqslant \Delta(T), \qquad (1)$$

where $\Delta(T)$ is the gap in the electron spectrum of the superconductor. For bulk superconductors, this condition can be satisfied in alloys and compounds with a small path length of the electrons, which assures large values of the Ginzburg-Landau parameter $\varkappa$ and of H_{c2}^{orb}. In this case, thanks to the different temperature dependences of H_{c2}^{orb} and Δ, condition (1) is satisfied only at temperatures that are remote from T_c.

A similar picture also exists for films in perpendicular magnetic fields, since the critical perpendicular magnetic field of the films $H_{c\perp} = H_{c2}$ (this equality is satisfied for films of type II superconductors of any thickness, and for sufficiently thin films of type I superconductors[6]), while condition (1) can be satisfied not only for films of alloys and compounds, but also for films of pure metals, thanks to the possibility of obtaining films with a very small path length of the electrons. So far as the parallel critical magnetic fields of films are concerned, their value and temperature dependence are such that the condition (1) for H_c^{orb} can be satisfied even near T_c [$H_{c\parallel}^{orb}$ is larger than H_c^{orb} by a factor of $\xi(T)/d$ is sufficiently thin films, where $\xi(T)$ is the temperature-dependent coherence length and d is the thickness of the film].

It should be remarked that satisfaction of condition (1) is facilitated at small Fermi velocities for the electrons v_F (and, consequently, small coherence lengths ξ_0), which are characteristic for transition metals, where values of v_F that are an order of magnitude smaller than in simple metals are possible.

In the present work, we have studied the dependence of H_c on the film thickness of two transition metals—vanadium and technetium—down to thickness of $d \simeq 60$ Å. The results are interpreted from the viewpoint of the effect of spin phenomena. In addition, since vanadium and technetium differ by about a factor of two in their atomic numbers, it is of interest to follow the effect of spin-orbit scattering,[4] the probability of which depends strongly on the atomic number.[7]

2. EXPERIMENTAL METHOD

For the present investigations, we used samples prepared by ion sputtering[8] in a superhigh vacuum setup, with the following partial pressures of the residual gases: hydrogen–$p < 10^{-9}$ Torr, other residual gases–$p < 10^{-10}$ Torr, and at a pressure of the working gas (krypton) 5×10^{-6} Torr. To remove the effects of the surrounding medium, and of the nonreproducibility of

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the properties of the samples, which could arise because of the ambiguity of the conditions on the boundaries of the film, the layer of metal was placed between thin (10–40 Å) carbon layers,[1)] which had excellent protective (mechanical and anticorrosive) properties.

Data characterizing the properties of the individual samples used in the present work were given in our previous papers on films of technetium[9] and vanadium.[10] The critical magnetic fields were measured by the resistance method, as is described in Refs. 9 and 10. Parallel orientation of the plane of the film relative to the direction of the field was established by rotating the sample about an axis perpendicular to the axis of the superconducting solenoid until the resistance was a minimum. The measurement current was perpendicular to the magnetic field. The remaining experimental details have been set forth in Refs. 9 and 10.

3. RESULTS

The transition curves $R(T)$ in a parallel magnetic field for films of both technetium and vanadium exhibited little change of shape and width over a rather wide range of fields near $H=0$. The width of the transition curves $R(H_{\parallel})$ changes upon change in the temperature. The decrease in the width of the transition curve $R(H_{\parallel})$ with lowering of the temperature is connected with the decrease in the steepness of the $H_{c\parallel}(T)$ plot. Examples of the transition curves are given in Fig. 1.

If we define $H_{c\parallel}$ in accord with the "beginning" or "end" of the transition curve $R(H_{\parallel})$ [i.e., in accord with the points of intersection of the continuations of the mean straight-line portion of the curve $R(H_{\parallel})$ with the lines $R=0$ and $R=R_n$, where R_n is the resistance in the normal state], we then obtain $H_{c\parallel}(T)$ curves shifted relative to each other by the constant amount δT. The choice of the criterion for the definition of $H_{c\parallel}$ leads only

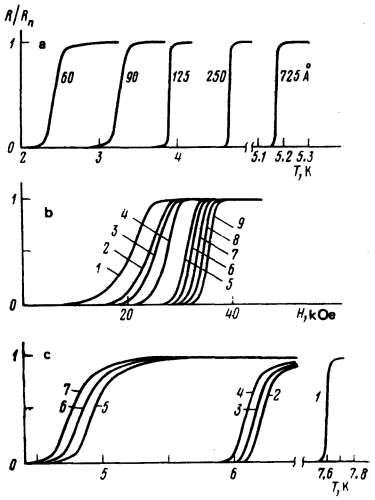

FIG. 1. Transition curves: a) vanadium film in zero magnetic zero magnetic field, the numbers on the curves indicate the thickness in Å; b) vanadium films ($d=60$ Å) in a field $H_{\parallel}$, temperature (in K): curve 1—2.18, 2—2.11; 3—2.96; 4—2.01; 5—1.86; 6—1.83; 7—1.76; 8—1.65; 9—1.52; c) in technetium films (Of thickness $d=1600$ Å at $H=0$ (curve 1); $d=80$ Å at $H=0$ (curve 2), 31.2 kOe (3) and 48.5 kOe (4); $d=55$ Å at $H=0$ (5), 33.9 kOe (6) and 50.8 kOe (7).

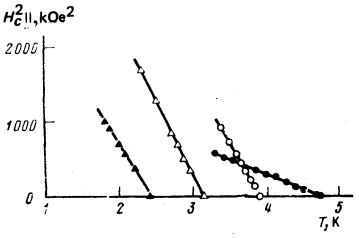

FIG. 2. Dependence of $H_{c\parallel}{}^2(t)$ of a vanadium film: ●—$d=250$ Å, ○—125 Å, △—88 Å, ▲—60 Å.

to a parallel shift in the $H_{c\parallel}(T)$ dependence. For definiteness, we choose as $H_{c\parallel}$ the field at which $R=R_n/2$.

A linear dependence of $H^2_{c\parallel}$ on the temperature is observed for all the investigated samples in a certain region close to T_c (Fig. 2). The observed length of the linear portion for the thickest films amounts to ~1.5K for $d=500$ Å, and $\leq 0.4K$ for $d\geq 1000$ Å. In accord with theory,[12] the size of this linear region is determined by the condition $2\xi(T)\gtrsim d$. For thicknesses 125 $\leq d\leq 500$ Å, the linear dependence is observed at temperatures that differ from T_c by about 1.5 K or less, For very thin films of vanadium, the region of linearity is somewhat reduced (down to ~0.3 K at $d\simeq 60$–70 Å). In the case of technetium films of thickness $d>110$ Å, the linearity is observed over the entire range of temperatures, the lower boundary of which, because of the large slope of H_c^2 relative to T, is determined by the maximal field at which the measurements were carried out (50–60 kOe).

The derivative $|dH^2_{c\parallel}/dT|_{T_c}$ was determined in the linear portion of $H_c^2(t)$ the values of which, as a function of the inverse thickness, are shown for vanadium in Fig. 3, and for technetium in Fig. 4. In both cases, in the range of large thicknesses, the quantity $|dH^2_{c\parallel}/dT|_{T_c}$ increases with increase in $1/d$, then reaches a maximum (at $d\simeq 100$ Å); for very thin films it tends to decrease. The values of $|dH^2_{c\parallel}/dT|_{T_c}$ range from 16 to 20 000 kOe²/K for technetium, and from 4 to 2 100 kOe²/K for vanadium.

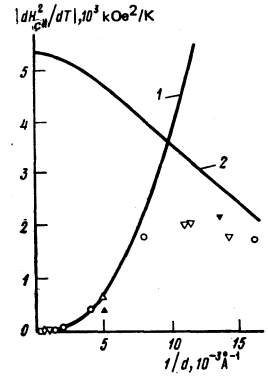

FIG. 3. Dependence of $|dH_{c\parallel}{}^2/dT|$ on the inverse thickness of a vanadium film (the different symbols, which denote the experimental points, correspond to different experiments on the preparation of the film): 1—theoretical curve, which takes into account only the orbital effects; 2—theoretical curve corresponding to the paramagnetic limit.

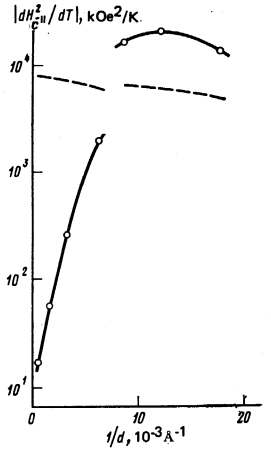

FIG. 4. Experimental dependence of $|dH_{c\parallel}{}^2/dT|$ on the inverse thickness of a technetium film (solid line). The dashed line is the paramagnetic limit. The break in the curve corresponds to the transition from the hcp to the fcc modification.

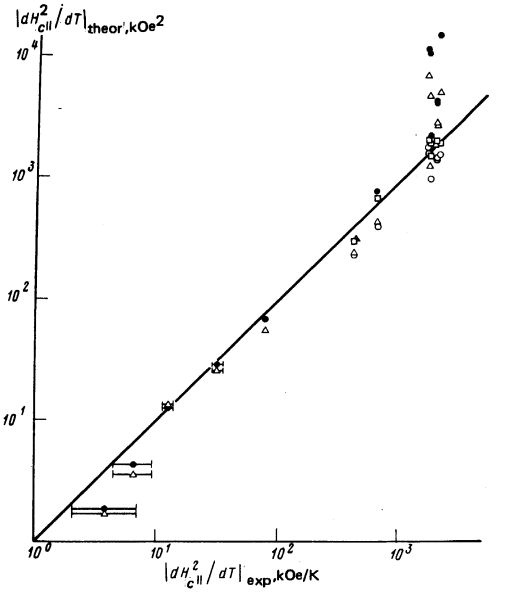

FIG. 5. Theoretical values of $|dH_{c\parallel}{}^2/dT|_{\text{theor}}$ relative to the experimental values $|dH_{c\parallel}{}^2/dT|_{\text{exp}}$ (in log-log plot) for vanadium film: △ and ● are the theoretical values, calculated without account of spin effects (i.e., $|dH_{c\parallel}{}^2/dT|_{\text{theor}} = |dH_{c\parallel}{}^{\text{orb}})^2/dT|$, ● and □ are the theoretical values calculated with account of both orbital and spin mechanisms of disruption of superconductivity by the magnetic field. $|dH_{c\parallel}{}^2/dT|_{\text{theor}}$ was calculated with account (○ and △) and without account (□ and ●) of the surface layers. The points ○ and □ in the region $|dH_{c\parallel}{}^2/dT| < 100$ kOe²/K ($d \geqslant 500$ Å) are not shown, since they coincide with △ and ●.

4. DISCUSSION

A. Vanadium films

It should first be noted that the observed discontinuance of the increase[2)] of $|dH_{c\parallel}^2/dT|_{T_c}$ upon decrease in the film thickness does not find its explanation within the framework of a theory which takes into account only the orbital effects.[12-14] For films of thickness $d = 60$–70 Å (both in the case of vanadium and technetium), the experimental values of $|dH_{c\parallel}^2/dT|_{\text{exp}}$ are lower than the theoretical $|d(H_{c\parallel}^{\text{orb}})^2/dT|_{\text{theor}}$, by a factor of 2.5–3. For vanadium films, this is seen from Fig. 5, on which (in logarithmic scale) are plotted the theoretical values of $|dH_{c\parallel}^2/dT|_{\text{theor}}$ relative to the experimental values $|dH_{c\parallel}^2/dT|_{\text{exp}}$. The theoretical values were computed both for the purely orbital case, and for the case of joint action of orbital and spin mechanisms (see below). The values corresponding to the orbital mechanism, $|dH_{c\parallel}^{\text{orb}})^2/dT|$, are found from the formula

$$\left|\frac{d(H_{c\parallel}^{\text{orb}})^2}{dT}\right| = \frac{6\varphi_0}{\pi}\frac{|dH_{c\perp}/dT|_{T_c}}{d^2}, \tag{2}$$

which is obtained if, in the expression[12,14]

$$H_{c\parallel}^{\text{orb}} = 3^{1/2}\varphi_0/\pi d\xi(T), \tag{3}$$

which is valid at $d \leqslant 2\xi(T)$, we replace $\xi(T)$ from

$$H_{c\perp} = \frac{\varphi_0}{2\pi\xi^2(T)}, \tag{4}$$

square the obtained for H_c^{orb}, and differentiate with respect to the temperature. In formulas (2)–(4), d is the film thickness, φ_0 is the magnetic flux quantum, $\xi(T)$ is the temperature-dependent coherence distance.

The theoretical values were calculated for two cases: it was assumed that a) the vanadium film is homogeneous in the direction perpendicular to its plane, and b) near the surface there exist nonsuperconducting layers with a high resistivity. Analysis of the aggregate of our data on the thickness dependence of the quantities T_c, $|dH_{c\perp}/dT|_{T_c}$, the residual resistivity ρ_m and the resistivity at room temperature ρ_{300} points to a probability of the existence of such surface layers of thickness 12–19 Å (independent of the thickness of the sample). The nature of these surface layers has not been determined at the present time. Similar surface layers apparently exist in films of other transitions metals, including technetium. Using the results of a theoretical paper of Usadel[15] and the parameters of the assumed surface layers in our experiments, we can draw the conclusion that allowance for these layers reduces to the replacement of the total thickness of the vanadium film d by a thickness that is less by twice the thickness of the surface layer.

Staying within the framework of the theory which takes into account only the orbital effects, we can attempt to explain the observed discrepancy between the experiment and theory by assuming that there exists an unaccounted for perpendicular component of the magnetic field, which would lead to a certain lowering of the critical field in comparison with the case of an ideal parallel orientation (local departures from parallelism can stem, for example, from the roughness of the substrate, while the effect of the roughness would be substantial only for rather thin films, with a depth of the roughness comparable with the thickness of the sample). Analysis of such a possibility was carried out by us on the basis of a formula of Tinkham:[6]

$$\frac{H_c(\varphi)\sin\varphi}{H_{c\perp}} + \left[\frac{H_c(\varphi)\cos\varphi}{H_{c\parallel}}\right]^2 = 1, \tag{5}$$

where φ is the angle between the direction of the field and the film surface. This analysis showed that the at-

tempt to choose an angle φ such that the experimental values of $|dH_c^2/dT_c|$ agree with the calculation from Eq. (6) leads to an entirely different temperature dependence of $H_c(T)$ than that observed. Thus it is not possible to attribute the observed results to an unaccounted for component.

We now proceed to a discussion of the results with account taken of the effect of spin phenomena. The microscopic theory which takes into account the effect of the magnetic field on the electron spin[5,16] along with orbital effects, leads to the following formula for $H_{c\parallel}^2$ in the region $T \gtrsim 0.7T_c$ (at $l \ll \xi_0$):

$$H_{c\parallel}^2(T) = 8kT \ln \frac{T}{T_c} \Big/ \left(\frac{\pi}{9} \frac{d^2 e^2 l v_F}{c^2 \hbar} + \frac{16.8 \mu_B^2}{\pi^2 kT} \right) \quad (6)$$

(where l is the k path length of the electrons, μ_B is the Bohr magneton, v_F is the Fermi velocity of the electrons) and, consequently, to

$$\left| \frac{dH_{c\parallel}^2}{dT} \right| = 8k \Big/ \left(\frac{\pi}{9} \frac{d^2 e^2 l v_F}{c^2 \hbar} + \frac{16.8 \mu_B^2}{\pi^2 k T_c} \right). \quad (7)$$

At $l_F d \to 0$ (i.e., in the case of smallness of the term corresponding to orbital effects in comparison with the term responsible for spin effects), it follows from this formula that the limiting slope $|dH_c^2/dT|$, which is due to the action of the field on the spin, is equal to

$$\left| \frac{dH_{c\parallel}^2}{dT} \right| = \frac{\pi^2}{2.1} \frac{k^2 T_c}{\mu_B^2}. \quad (8)$$

The curve corresponding to this "paramagnetic limit," is shown in Fig. 3 (curve 2). We have drawn on this drawing curve 1 constructed with account taken of only orbital effects. Curves 1 and 2 are smoothed curves passing through points corresponding to the theoretical values obtained from formulas (2) or (8), with use of the experimental values of $|dH_{c\perp}/dT|_{T_c}$ and T_c. The points themselves are not shown in Fig. 3 so as not to clutter up the drawing.

The intersection of curves 1 and 2 takes place at a thickness $d \approx 100$ Å. Thus, in the region $d > 100$ Å, the orbital mechanism of disruption of the superconductivity by a parallel magnetic field dominates, while in the region $d < 100$ Å, it is the spin (paramagnetic) mechanism. The fact that the growth of $|dH_{c\parallel}^2/dT|$ with decrease in the thickness ceases just at the thickness 100 Å indicates that the cessation of increase in $|dH_{c\parallel}^2/dT|$ is due to the paramagnetic effect.

We now turn to Fig. 5 and give our attention to those points which show a comparison of the experimental data with the theory, taking into account both mechanisms—orbital and spin. In this case, $|dH_{c\perp}^2/dT|_{\text{theor}}$ has been calculated from formula (7), in which we have substituted the value of lv_F found from the data for the perpendicular critical field, according to the formula

$$lv_F = \frac{12}{\pi} \frac{ck}{e} \Big/ \left| \frac{dH_{c\perp}}{dT} \right|_{T_c}, \quad (9)$$

which follows from the work of Gor'kov[17] with account of the fact that $H_{c\perp} = H_{c2}$.[6] The calculation in accord formula (7) is carried out only for $d \leq 125$ Å ($|dH_{c\parallel}^2/dT| > 10^2$ kOe^2 K, $l/\xi_0 < 0.07$). For $d > 125$ Å, the second term in the denominator of the right side of formula (7), which describes the effect of the paramagnetic phenomenon, can be neglected in comparison with the first. The relation between the first and second terms depends principally on the value of d^2. Consequently, in the region $d > 125$ Å, we need take into account only the orbital effects. The calculations for this region have been carried out according to formula (1).

It is seen that account of the effect of the paramagnetic phenomenon leads to satisfactory agreement of experiment with theory over the entire range of thicknesses, including the region $d \lesssim 100$ Å ($|dH_{c\parallel}^2/dT| > 10^3$ kOe^2/K). It must be noted that somewhat better agreement is obtained for the model of a film with surface layers than for the model of a film that is homogeneous over its thickness.

B. Technetium films

In Fig. 4, along with the experimental points, the graph of the dependence of $|dH_{c\parallel}^2/dT|$ on $1/d$ for technetium films shows also the theoretical curve, corresponding to the paramagnetic limit, in accord with formula (9). The break in this curve in the region $1/d$, which corresponds to the thickness 110–160 A, is due to the structural transition of the technetium film from the hexagonal close packed (hcp) to the face-centered cubic (fcc) phase.[9] Just as in vanadium films, the increase in $|dH_{c\parallel}^2/dT|$ with decrease in the thickness ceases at $d \leq 110$ Å. The difference from the case of the vanadium film is that the value of $H_{c\parallel}$ in the region $d \leq 110$ Å is several times greater than the paramagnetic limit calculated from formula (8). For films $d = 80$ and 55 Å, this excess reaches threefold. Qualitatively, this result can be explained by the effect of spin-orbit scattering.

Spin-orbit scattering (in contrast to scattering from magnetic impurities) does not lead to a disruption of the Cooper pairs, but increases the spin susceptibility of the superconductor.[7,18,5] Thanks to this, the spin-orbit scattering decreases the free energy of the superconducting state in a magnetic field, increasing the difference of the free energies of the normal and superconducting states. Thus, the effect of spin-orbit scattering is opposed to the paramagnetic effect, which lowers the free energy of the normal state and thereby decreases the difference of the free energies of the normal and superconducting states. The fact that the appearance of a spin-orbit effect has not been observed in vanadium films, while it has been found in technetium films is in qualitative agreement with the assertion that the role of spin-orbit scattering increases sharply with increase in the number of the element in the periodic table[7] (the atomic number of vanadium is 23, of technetium, 43).

According to Abrikosov and Gor'kov,[7] the probability of spin flip in each set of scattering of the electron is equal, in order of magnitude, to

$$\frac{\tau_{tr}}{\tau_{so}} = \left(\frac{Ze^2}{\hbar c} \right)^4, \quad (10)$$

where τ_{tr} is the transport relaxation time, τ_{so} is the time between spin flips, Z is the nuclear charge. This relation agrees with the results of Crow *et al.*,[4] who measured the parallel critical fields of thin films of Al, Sn and Ga, and who determined the values of τ_{so} from

a comparison with the theory which takes into account the paramagnetic and spin-orbita phenomena.[5] Meservey and Tedrow,[3] using measurements not only of the critical magnetic field, but also of the tunnel effect for the determination of τ_{so}, did not find quantitative agreement with formula (10), but they also observed a strong increase of τ_{tr}/τ_{so} with increase in Z. An excess over the paramagnetic limit (approximately threefold) was observed by Bogomolov *et al.*[19] in the measurement of the critical fields of thin threadlike specimens of mercury in the case of parallel orientation of the threads relative to the field. A strong excess over the paramagnetic limit was found in measurements of $H_{c\perp}$ of ultrathin films of bismuth.[2] Critical fields with a smaller excess over the paramagnetic limit have also been observed by other authors, both in bulk samples and in films.[20,21]

The degree of effect of spin-orbit scattering on the value of the critical magnetic field can be characterized by the parameter $b=\hbar/3\tau_{so}\Delta(0)$ where $\Delta(0)$ is the energy gap at $T=0$. By finding τ_{so} from (10), and τ_{tr} from resistance data, we obtain that for a film of technetium of thickness $d\lesssim 100$ Å the parameter b has an appreciable value ($b_{T_c}=1.5-1.7$), while for vanadium films this parameter is small at all those thicknesses ($b_V \lesssim 4\times 10^{-2}$).

Thus, the observed dependences of $|dH_{c\parallel}^2/dT|$ on thickness in the region of ultrathin films of vanadium and technetium are explained by the effect of paramagnetic phenomenon. But whereas the results for vanadium films agree quantitatively with the theory which takes into account the effect of spin paramagnetism along with the usual mechanism of disruption of superconductivity in a magnetic field (which is connected with a change in the orbital motion of the electrons under the action of the field), in technetium films a threefold excess over the paramagnetic limit is reached and can be qualitatively explained as due to the effect of spin-orbital scattering.

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