Critical magnetic field of NbC: new data on clean superconductor films

S. I. Krasnosvobodtsev, N. P. Shabanova, E. V. Ekimov, V. S. Nozdrin, and E. V. Pechen'

P. N. Lebedev Physics Institute, Russian Academy of Sciences, 117924 Moscow, Russia
(Submitted 10 March 1995)
Zh. Éksp. Teor. Fiz. **108**, 970–976 (September 1995)

The temperature dependence of the upper critical magnetic fields of exceptionally low-defect-density films of the superconducting compound NbC has been investigated, and previously unknown parameters of this clean superconductor and its electronic characteristics have been evaluated. An electron density of states at the Fermi level equal to 1.3 states/eV·Nb atom, a Fermi velocity equal to 2.2×10^7 cm/s, a plasma frequency equal to 3.6 eV, and a coherence length $\xi_0 = 24$ nm have been obtained with an electron mean free path exceeding 40 nm. A vortex-free state existing over the entire temperature range below T_c, which causes a many-fold increase in the critical magnetic field of the films when the field is aligned parallel to their surface, has been discovered in very thin films of superconducting niobium carbide.

1. INTRODUCTION

Niobium carbide has been an object of intent interest for a long time owing to its numerous unusual properties.[1,2] However, the extensive list of publications on NbC includes few that were devoted to the study of the upper critical magnetic field of this superconducting compound. An investigation of the upper critical field of a type-II superconductor makes it possible to obtain a whole set of electronic characteristics of the material, such as the electron density of states at the Fermi level, the Fermi velocity, the coherence length, the electron mean free path, etc.[3–9] Either the literature does not offer any data on some of these parameters for niobium carbide, or they pertain to samples of nonstoichiometric composition. Taking into account that the properties of NbC are highly sensitive to the defect density in the crystal structure, it seems important to determine its electronic characteristics in samples with a nearly stoichiometric composition. Obtaining such data for very thin films of NbC is of special interest from the standpoint of the prospects of their application.

2. EXPERIMENT

Niobium carbide films synthesized by reactive laser deposition similar to those described in Ref. 10 were investigated in this work. This method has been used successfully to obtain NbC samples characterized by the most nearly stoichiometric composition and the highest superconducting transition temperature for this compound ($T_c = 12$ K). The employment of plates of single-crystal sapphire with the $(\bar{1}012)$ orientation in the present work, as well as the careful selection of the substrate temperature and measures to ensure its stability throughout the deposition process, made it possible to significantly lower the concentration of defects that are not carbon vacancies. The normal-state resistivity of the most perfect films at $T = 13$ K was equal to about 4 $\mu\Omega\cdot$cm, which is an order of magnitude below the presently known values, including those for single-crystal samples.[1,2]

The thickness of the films d was measured on an interference microscope in the visible wavelength range and determined from the interference of the incident x radiation and the radiation reflected at small angles. The resistivity was measured by the four-point probe technique. The magnetic field was created by a superconducting solenoid.

The temperature dependence of the upper critical field was determined from the temperature shift of the superconducting transition caused by a magnetic field. Sharp resistivity transitions, which broaden slightly in a magnetic field and are characteristic of homogeneous structures with good flux pinning, were observed. The temperature of the transitions was determined at the level of 0.5 of the normal-state resistivity at 13 K. The error due to the broadening amounted to 1 to 15% for different samples. The results of the measurement of the upper critical magnetic field for the NbC films are presented in Fig. 1.

3. UPPER CRITICAL FIELD $H_{c2}(T)$ AND ELECTRONIC CHARACTERISTICS OF NBC FILMS

The data obtained in a magnetic field perpendicular to the film (Fig. 1a) can be interpreted as the temperature dependence of the upper critical field $H_{c2}(T)$ (Ref. 11). Its linear character, which was observed over a broad temperature range near T_c for NbC films, corresponds to the theoretical temperature dependence of the Ginzburg–Landau coherence length

$$\xi(T) = \xi(0)(1 - T/T_c)^{-1/2}.$$

Taking into account the relation

$$H_{c2}(T) = \Phi_0 / 2\pi\xi^2(T),$$

where Φ_0 is the magnetic flux quantum and the parameter $\xi(0)$ of the sample can be evaluated from the slope $-dH_{c2}/dT$ of the measured dependence of $H_{c2}(T)$:

$$-\frac{dH_{c2}}{dT} = \frac{\Phi_0}{2\pi T_c \xi^2(0)}. \quad (1)$$

 1063-7761/95/090534-04$10.00

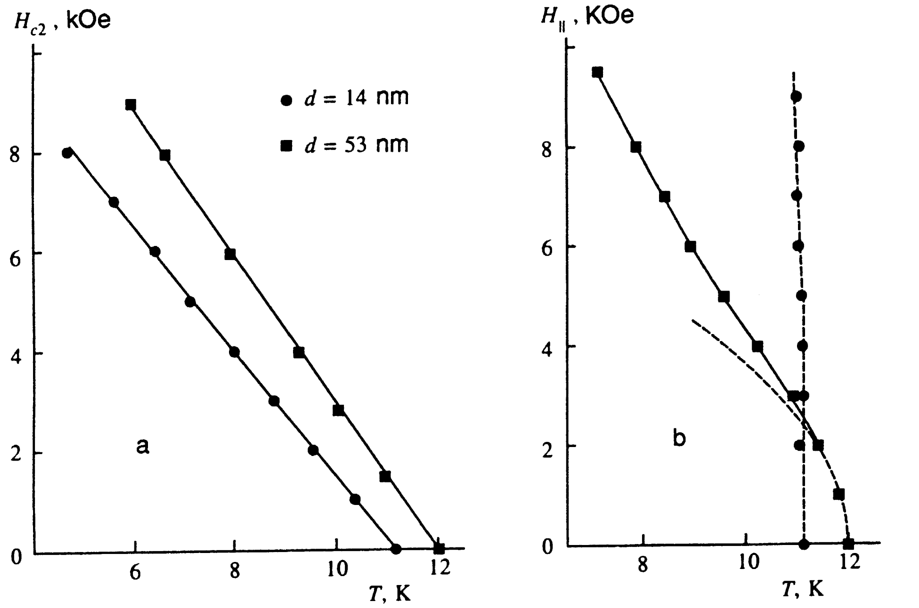

FIG. 1. Temperature dependence of the upper critical field of NbC films of various thickness measured in magnetic fields perpendicular (a) and parallel (b) to their plane. The theoretical curves obtained from Eq. (5) are shown as dashed lines.

As follows from the Gor'kov equations[12] and the relations of the Ginzburg–Landau–Abrikosov–Gor'kov (GLAG) theory for type-II superconductors,[3–9] the scattering of electrons on defects in a structure causes variation of its upper critical field. If the electron density of states at the Fermi level $N(0)$ remains unchanged, this results in an almost linear increase in $-dH_{c2}/dT$ with the normal-state resistivity ρ (Refs. 3 and 9).

A similar dependence is discovered for NbC films (Fig. 2). The electronic characteristics of this material can be evaluated from its parameters.

Extrapolation of such a dependence to $\rho=0$ gives the value of the slope $-dH^0_{c2}/dT$ (Refs. 6 and 9) of the clean superconductor, where the electron mean free path l is much greater than the coherence length.

The empirical value $H^0_{c2}(4.2)=7$ kOe and the critical field of the films were considerably lower than the known values for the previously investigated single crystals, which were classified as "dirty" superconductors[13] owing to their relatively high resistivity.

$$\xi(0) = 0.74\xi^*_0.$$

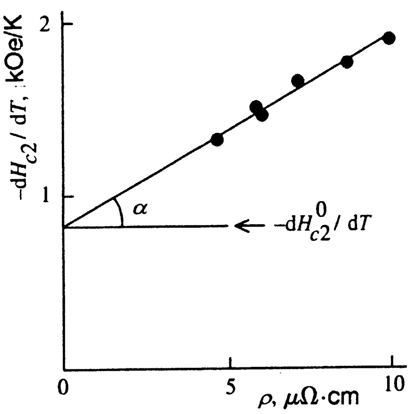

FIG. 2. Dependence of the slope $-dH_{c2}/dT$ of the temperature dependence of the upper critical magnetic field near T_c on the resistivity of NbC films.

After evaluating $\xi(0)$ for clean NbC from the expression (1) for $-dH^0_{c2}/dT$, the coherence length ξ^*_0 appearing in the microscopic theory can be found for it. In the clean limit $l \gg \xi^*_0$, these parameters are related by the expression

$$\xi(0)=0.74\xi^*_0.$$

It follows from an analysis of the expressions of the GLAG theory[9] that the following relation holds with good accuracy when $l \gg \xi^*_0$:

$$\frac{dH_{c2}/dT - dH^0_{c2}/dT}{dH^0_{c2}/dT} = \frac{0.9\xi^*_0}{l}. \tag{2}$$

Using the data obtained (Fig. 2), the value of the ξ^*_0/l for each sample can be obtained from (2), and then, knowing the coherence length, we can determine the distance l from the limiting value of the slope. The mean free path in some of the films investigated exceeds 40 nm when $\xi^*_0=24$ nm, allowing us to regard them as clean superconductors. Estimates of $-dH^0_{c2}/dT$, ξ^*_0, and l were obtained for samples with thicknesses greater than 50 nm (the dimensional effect in the conductivity[14] becomes significant for thinner films).

As was shown in Ref. 7, the most important factor determining the value of the critical field of a clean superconductor is the square of the Fermi velocity averaged over the Fermi surface $\langle v_F^2\rangle$. From the expressions of the GLAG theory for a clean superconductor with a complex Fermi surface[15] and the parameter v_F^* renormalized with consideration of the strong coupling we have[8,9]

$$-\frac{dH^0_{c2}}{dT} = 3.17\times 10^{16}\eta \frac{T_c}{\langle v_F^{*2}\rangle}. \tag{3}$$

Hence, with the correction for strong coupling $\eta=1.05$, which was calculated on the basis of the data from tunneling measurements of NbC films,[16] we obtain the estimate $\langle v_F^{*2}\rangle^{1/2}=2.2\times 10^7$ cm/s.

Generalization of the relation

$$\frac{dH_{c2}}{dT}\sim N^*(0)\rho,$$

which is valid in the "dirty" limit $l\ll\xi_0^*$ (Ref. 3), leads to a similar expression for sufficiently clean superconductors

$$\frac{dH_{c2}}{dT}-\frac{dH_{c2}^0}{dT}\sim N^*(0)\rho,$$

and the renormalized electron density of states $N^*(0)$ is determined from the tangent of the angle α (Ref. 9) (Fig. 2). The expression

$$N^*(0)=3.04\cdot 10^{26}\,\frac{1}{\eta}\,\frac{|dH_{c2}/dT-dH_{c2}^0/dT|}{\rho}, \tag{4}$$

works with good accuracy under the condition $l\geqslant\xi_0^*$, which holds for the films investigated. The relations were written for $-dH_{c2}/dT$ in Oe/K, for ρ in $\Omega\cdot cm$, and for $N^*(0)$ in states/erg$\cdot$cm^3 (per pair of spins). Hence, for NbC we have $N^*(0)=1.3$ states/eV$\cdot$Nb atom. The corresponding value of the coefficient in front of the electronic specific heat is equal to 0.22 mJ/cm$^3\cdot$K^2 and is consistent with the data obtained from measurements of the specific heat[17] of bulk samples with a nearly stoichiometric composition.

Using the value $\lambda=0.9$ of the electron–phonon coupling constant for NbC, which was obtained from tunneling data,[16] we can evaluate the electronic band characteristics:

$$v_F=v_F^*(1+\lambda),\quad N(0)=\frac{N^*(0)}{1+\lambda}.$$

The corresponding value of $\langle v_F^2\rangle^{1/2}$ is equal to about 4×10^7 cm/s, and $N(0)\simeq 0.7$ states/eV$\cdot$Nb atom. The resultant value $\omega_p=3.6$ eV of the low-temperature plasma frequency $\omega_p=(4\pi e^2\langle v_F^2\rangle N(0)/3)^{1/2}$, which has scarcely been studied for this compound, is of interest.

The error in the evaluations may be as high as 25%. The accuracy can be improved by expanding the experimental statistics.

4. THIN FILMS OF THE SUPERCONDUCTOR NBC IN A PARALLEL MAGNETIC FIELD

Among the values presented for the electronic characteristics of the NbC films investigated, the high value of the electron mean free path, which is unusual for superconducting compounds, deserves attention. Together with the fairly high coherence length, it makes it possible to realize the conditions of a vortex-free state[18–21] in films of relatively large thickness. The condition $d<1.8\xi(T)$ (Refs. 18 and 21) practically precludes the observation of this effect for most superconducting alloys and compounds having values of l and $\xi(0)$ of the order of several atomic layers.

NbC films in a parallel magnetic field (Fig. 1b) exhibit a temperature dependence of the critical field which is characteristic of the vortex-free state near T_c, at which $\xi(T)$ is

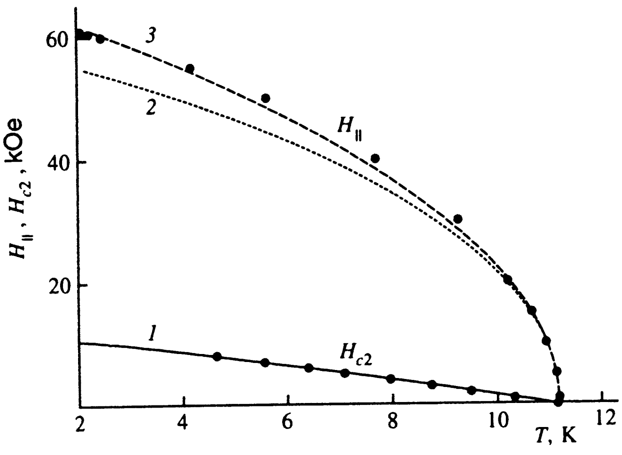

FIG. 3. Temperature dependence of the upper critical field of a thin NbC film with a thickness $d=14$ nm in perpendicular (H_{c2}) and parallel ($H_\parallel$) magnetic fields. The points indicate measured values. *1*—plot of $H_{c2}(T)$ from the GLAG theory; *2*—theoretical curve in the Abrikosov model;[19] *3*—theoretical curve in the Shapoval model[20] [Eq. (5)].

considerably greater than the thickness of the film, already when $d=53$ nm. When the temperature was lowered to a level at which the quantity $2\xi(T)$, which characterizes the size of a vortex, becomes comparable to the film thickness, the function $H_\parallel(T)$ underwent a change in character, which corresponds to the penetration of vortices into the sample[18,21] (Fig. 1b). It was accompanied by an abrupt change in the broadening of the superconducting transitions, which is attributed to the dissipative motion of the vortex system. The resultant value of the penetration temperature $T\simeq 10$ K is consistent with the theoretical value.

For a thinner film with $d=14$ nm and $\xi(0)=15$ nm, the criterion $d<1.8\xi(T)$ holds over the entire temperature range below T_c, so that vortices cannot penetrate at any temperature. Behavior of the function $H_\parallel(T)$ consistent with this was observed experimentally (Fig. 3).

The abrupt increase in the critical field observed as the thickness of the samples decreases corresponds to the theoretical dependence $H_\parallel\propto d^{-3/2}$ (Ref. 20).

According to the results of Shapoval's work,[20] which gave a very detailed and exact solution of the problem of the critical field of a thin film in the vortex-free state when there is diffuse electron scattering on the surfaces, the value of $H_\parallel(T)$ near T_c is specified by the relation

$$H_\parallel(T)=0.86\,\frac{\Phi_0}{(d^3\xi_0^*)^{1/2}}\left(1-\frac{T}{T_c}\right)^{1/2}. \tag{5}$$

Substitution of the values of d and ξ_0^* found above into this expression leads to good quantitative agreement between the theoretical plots of $H_\parallel(T)$ and the experimental data (Fig. 1b, curve *3*). This confirms the reliability of the values obtained for the electronic characteristics of low-defect-density NbC films with a nearly stoichiometric composition.

In the case of a thick film, relation (5), which should hold when $d\ll\xi(T)$, was found to be valid over the entire temperature range. In fact, it follows from Abrikosov's calculations[19] that in the case $d \leqslant \xi(T)$ the temperature dependence of the parallel critical field maintains a similar character (Fig. 3). Quantitative agreement with experiment is achieved in the Abrikosov model to within the experimental error when the parameters d and $\xi(0)$ are varied.

5. CONCLUSIONS

Thus, the upper critical magnetic field of niobium carbide films with a low concentration of carbon vacancies and other defects has been measured in the present work. It has been shown that, unlike the previously investigated samples, they are practically clean superconductors.

The resistivity dependence of the upper critical magnetic field H_{c2} of the films has been used to evaluate the following renormalized electronic characteristics: $N^*(0)=1.3$ states/eV·Nb atom, $\langle v_F^{*2}\rangle^{1/2}=2.2\times 10^7$ cm/s, and $\omega_p=3.6$ eV. Extrapolation to the clean limit gives $H_{c2}^0(4.2)=7$ kOe and $\xi_0^*=24$ nm. The electron mean free path for the samples with a minimal number of defects typically exceeded 40 nm.

The values found for the electronic characteristics have made it possible to quantitatively describe the observed temperature dependence of the parallel critical magnetic field of NbC thin films in the vortex-free state on the basis of the theoretical calculations of Abrikosov and Shapoval.

The fabrication of NbC thin films with a large electron mean free path (of the order of 100 lattice constants) makes it one of the few superconducting compounds in which a vortex-free state can be produced over a broad temperature range.

This work was performed with support from the Interdisciplinary Scientific-Technical Program on High-T_c Superconductors (Project No. 93069).

[1] L. E. Toth, *Transition Metal Carbides and Nitrides*, Academic Press, New York, 1971 (Russ. transl. Mir, Moscow, 1974).
[2] G. V. Samsonov, G. Sh. Upadkhaya, and V. S. Neshpor, *Physical Materials Science of Carbides* [in Russian], Naukova Dumka, Kiev, 1974.
[3] H. Weismann, M. Gurvitch, A. K. Ghosh *et al.*, Phys. Rev. B **17**, 122 (1978).
[4] T. P. Orlando, E. J. McNiff, Jr., S. Foner, and M. R. Beasley, Phys. Rev. B **19**, 4545 (1979).
[5] A. I. Golovashkin, E. V. Pechen', and N. P. Shabanova, Zh. Éksp. Teor. Fiz. **82**, 850 (1982) [Sov. Phys. JETP **55**, 503 (1982)].
[6] R. E. Somekh, C.-G. Cui, and J. E. Evetts, J. Low Temp. Phys. **50**, 33 (1983).
[7] W. H. Butler, Phys. Rev. Lett. **44**, 1516 (1980).
[8] B. J. Dalrymple and D. E. Prober, J. Low Temp. Phys. **56**, 545 (1984).
[9] A. I. Golovashkin and N. P. Shabanova, Physica C **185–189**, 2709 (1991); Preprint P. N. Lebedev Physics Institute, Academy of Sciences of the SSSR, No. 39, 1991.
[10] A. I. Golovashkin, B. G. Zhurkin, A. L. Karuzskiĭ *et al.*, Fiz. Tverd. Tela (Leningrad) **28**, 3342 (1986 [Sov. Phys. Solid State **28**, 1881 (1986)]).
[11] M. Tinkham, Phys. Rev. **129**, 2413 (1963).
[12] L. P. Gor'kov, Zh. Éksp. Teor. Fiz. **37**, 1407 (1959) [Sov. Phys. JETP **10**, 998 (1960)].
[13] H. J. Fink, A. C. Thorsen, E. Parker *et al.*, Phys. Rev. **138A**, 1170 (1965).
[14] Yu. F. Komnik, *Physics of Metal Films* [in Russian], Atomizdat, Moscow, 1979.
[15] L. P. Gor'kov and T. K. Melik-Barkhudarov, Zh. Éksp. Teor. Fiz. **45**, 1493 (1963) [Sov. Phys. JETP **18**, 1031 (1964)].
[16] E. V. Pechen, S. I. Krasnosvobodtsev, N. P. Shabanova *et al.*, Physica C **235–240**, 2511 (1994).
[17] L. E. Toth, M. Ishikawa, and Y. A. Chang, Acta Metall. **16**, 1183 (1968).
[18] D. Saint-James and P. G. de Gennes, Phys. Lett. **7**, 306 (1963).
[19] A. A. Abrikosov, Zh. Éksp. Teor. Fiz. **47**, 720 (1964) [Sov. Phys. JETP **20**, 480 (1965)].
[20] E. A. Shapoval, Zh. Éksp. Teor. Fiz. **49**, 930 (1965) [Sov. Phys. JETP **22**, 647 (1966)].
[21] H. J. Fink, Phys. Rev. **177**, 732 (1969).

Translated by P. Shelnitz