Eur. Phys. J. B **18**, 397–399 (2000)

THE EUROPEAN
PHYSICAL JOURNAL B
EDP Sciences

Change in the chemical potential of niobium at transition to the superconducting state

V.I. Nizhankovskii

International Laboratory of High Magnetic Fields and Low Temperatures, Gajowicka 95, 53-421 Wroclaw, Poland

Received 10 April 2000

Abstract. A temperature-modulation technique was applied to investigate change in the chemical potential of niobium film at transition to the superconducting state. The first derivative of the chemical potential is continuous at T_c whereas the second derivative shows a jump.

PACS. 74.25.Bt Thermodynamic properties – 74.76.-w Superconducting films

1 Introduction

So far only three experiments on change in the chemical potential μ of electrons at the superconducting transition have been performed. The earliest two were unsuccessful: the experiment with temperature gradients along the niobium sample [1] was inadequate. In the experiment with tin [2] the sensitivity was insufficient. First definite results were obtained by the vibrating Kelvin probe method for high-T_c films of $YBa_2Cu_3O_{7-\delta}$ [3]. The authors of this work claimed to have observed a discontinuous change in slope of $\mu(T)$ at T_c. This result is in strong contradiction with the classical theory of superconductivity in which the transition to the superconducting state is the second order, *i.e.* the slope of $\mu(T)$ should be continuous at T_c. We think that the interpretation of experimental results in this work was rather tendentious. One can see that experimental curves ([3], Fig. 1) looks rather smooth.

In the present work we have measured the first derivative of the chemical potential of niobium film directly. For this purpose a temperature-modulation technique was used. We show that the first derivative is continuous and the second derivative has a jump at transition temperature.

2 Experimental

The method to measure the change in the chemical potential was based on determination of change in charge on the measuring capacitor consisting of the sample under investigation (niobium film) and the reference electrode (gold film). In order to obtain high sensitivity the capacitance of the measuring capacitor should be as large as possible. Figure 1 shows construction of our measuring capacitor. Niobium film (150 nm) was evaporated on silicon substrate (1 cm × 1 cm × 0.3 mm). SiO_2 layer (1000 nm) was used as an insulator. Gold (400 nm) was evaporated as a reference electrode and as an electrical contact to niobium film. All films were made by magnetron sputtering.

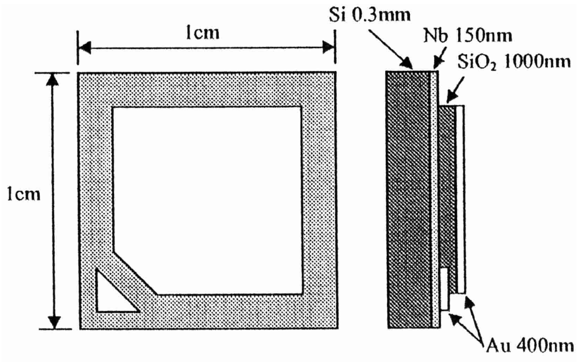

Fig. 1. The construction of the measuring capacitor.

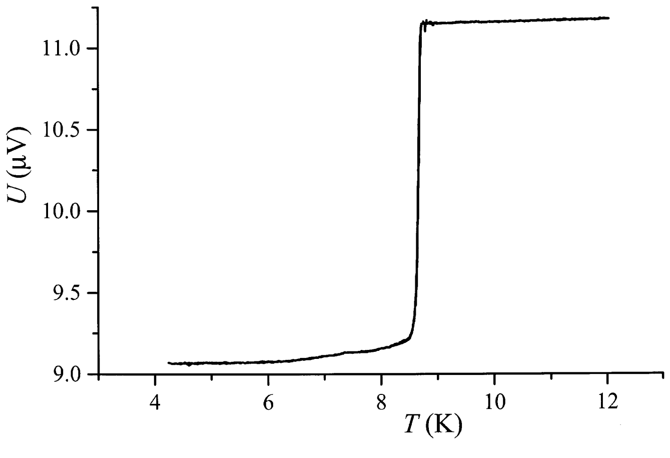

Fig. 2. The superconducting transition of niobium film.

In Figure 2 the superconducting transition of niobium film is shown. The transition was registered by an inductive method in the same experimental apparatus that was

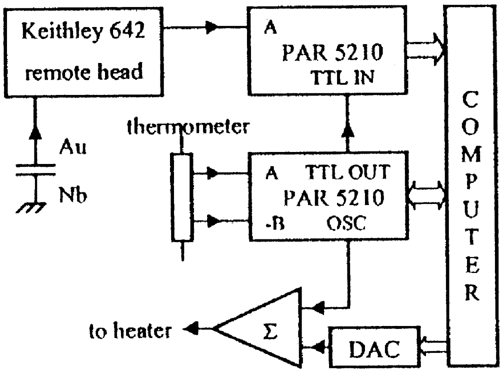

Fig. 3. Schematic diagram of the apparatus installation for measuring dμ/dT.

used to measure dμ/dT. For this purpose a modulating coil (7 turns) and a pickup coil (15 turns) were wound round the sample perimeter. Measurement was done at frequency 1237 Hz, modulating current was 4 mA. Superconducting transition begins at $T = 8.68$ K and is very narrow (about 0.05 K for 90% change of the signal).

The apparatus for dμ/dT measurements was based on a construction used for investigation the effect of a magnetic field on the chemical potential [4]. The sample was glued by silver paint to a polished copper block on which the heater and a germanium thermometer (Lake Shore) were mounted. The niobium film was grounded and the gold film was connected by a coaxial cable to the remote head of a Keithley 642 electrometer. Sapphire insulators were used in construction of the coaxial cable. In order to diminish the temperature gradient an additional heater was wound round the lower end of coaxial cable. The sample, gold film and central wire of coaxial cable were perfectly shielded from heaters and thermometer, because even very small parasitic capacitance between them results in erroneous signal at electrometer output. The coaxial cable and copper block were surrounded by a stainless steel, thin-wall tube, filled with gaseous helium as a heat exchanger (30 mm Hg at room temperature). This device was inserted into a liquid He storage vessel.

A schematic diagram of the electronics for the temperature-modulation technique of measuring dμ/dT is depicted in Figure 3. Two synchronized lock-in amplifiers (PAR 5210) were used: one for producing and measuring variation in temperature $\delta T = A_T \sin(\omega t)$ and another for measuring electrometer output $\delta Q = A_Q \sin(\omega t + \varphi)$. The temperature derivative of charge is given by dQ/dT = $(A_Q/A_T)\cos\varphi$. The temperature was modulated at a frequency of 0.555 Hz with amplitude $A_T = 0.15$ K. Measurements were taken between 4.5 and 19 K at intervals of 0.07 K. One run took about 15 hours.

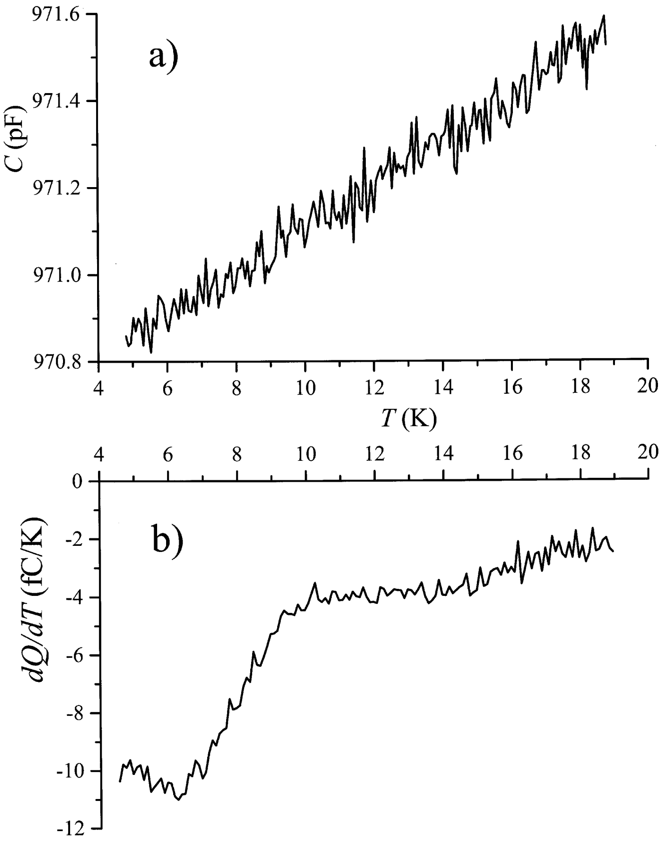

Fig. 4. Temperature dependences of capacitance of the measuring capacitor (a) and dQ/dT (b).

The results of the measurements are presented if Figure 4. Charge on the measuring capacitor Q ($Q = (\mu_{\mathrm{Nb}} - \mu_{\mathrm{Au}})C/e$, where e is the electron charge), changes with temperature due to change in the chemical potentials of the electrodes and due to change in the capacitance C itself (as it is well known [5] any intervening metals have no influence on Q). At the top of Figure 4 the temperature dependence of capacitance is depicted. Experimental points were obtained at fixed temperatures by measuring the charge on the measuring capacitor while a linearly changing voltage was applied to niobium film. At the bottom of Figure 4 the temperature dependence of dQ/dT is shown. This curve is the average of five runs (peak-to peak value of charge noise equals to 500e).

In order to obtain the temperature dependence of the chemical potential presented at the top of Figure 5 we have integrated the curve dQ/dT and divided the result by $C(T)$ (of course, the absolute position of the curve along the ordinate axis is arbitrary). The temperature derivative of the chemical potential presented at the bottom of Figure 5 was obtained by numerical differentiating the above curve. Because the change in capacitance with temperature did not exceed 0.1% the temperature derivative of the chemical potential is practically identical to dQ/dT (smaller noise results from smoothing applied with numerical differentiating).

3 Discussion

Our main result is to demonstrate the continuity of the first derivative of the chemical potential of niobium at transition to the superconducting state. In the vicinity

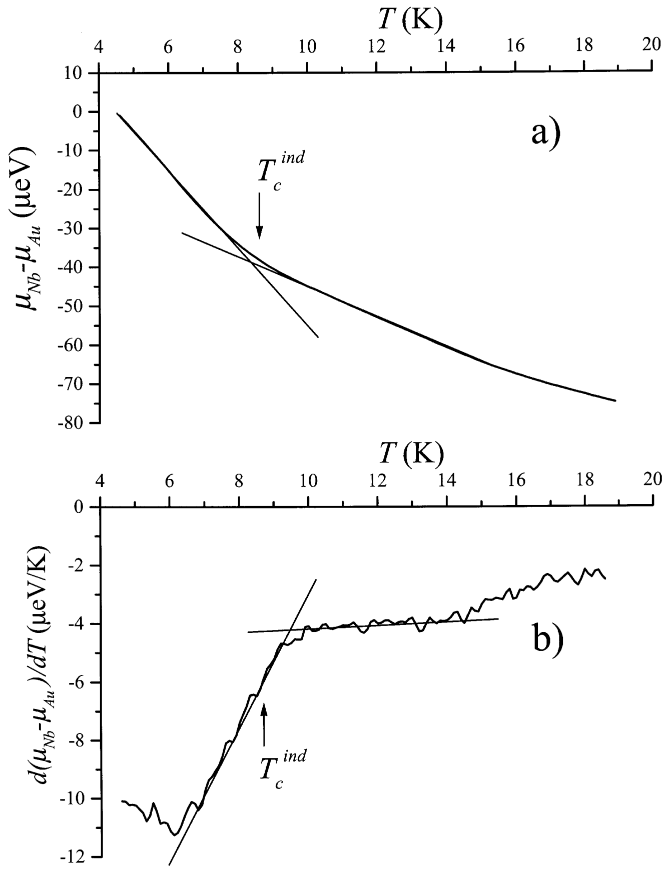

Fig. 5. Temperature dependences of the chemical potential difference (a) and its derivation (b). Arrows signed by $T_\mathrm{c}^\mathrm{ind}$ show position of the superconducting transition registered by the inductive method.

of the transition the first derivative of the chemical potential may be extrapolated by two straight lines (Fig. 5b). The jump of the second derivative of the chemical potential is in agreement with classical theories of the superconducting transition. It is interesting that the jump occurs at $T = 9.6$ K that is 0.9 K above $T_\mathrm{c}^\mathrm{ind}$ determined from inductive measurement. Moreover it is 0.4 K higher than the critical temperature of bulk niobium samples. We think that this experimental fact may be explained by enhanced superconductivity at the metal-dielectric interface [6,7]. Indeed, measurements of the chemical potential give information about very thin surface layer of a metal (of order the Debye screening length).

It is interesting that the temperature dependence of the chemical potential itself (Fig. 5a) may be approximated by two straight lines intersecting near $T_\mathrm{c}^\mathrm{ind}$. This result is qualitatively the same as obtained in work [3] for $\mathrm{YBa_2Cu_3O_{7-\delta}}$ where it was the basis for the conclusion about discontinuity of $\mathrm{d}\mu/\mathrm{d}T$. But this is obviously only an illusion (at least in the case of niobium).

The author is grateful to Dr. P. Dmitriev for preparing the sample and to Prof. J. Czerwonko for fruitful discussions of the problem.

References

1. P.B. Pipes, D.H. Darling, *Proc. 14th Intern. Conf. Low Temperature Physics, Otaniemi, 1975* (North-Holland/American Elsevier, 1975) vol. 2, p. 521.
2. M. Shott, A.J. Walton, Phys. Lett. A **60**, 53 (1977).
3. G. Rietveld, N.Y. Chen, D. van der Marel, Phys. Rev. Lett. **69**, 2578 (1992).
4. V.I. Nizhankovskii, S.G. Zybtsev, Phys. Rev. B **50**, 1111 (1994).
5. L.H. Fisher, R.N. Varney, Am. J. Phys. **44**, 464 (1976).
6. V.L. Ginzburg, Phys. Lett. **13**, 101 (1964).
7. D. Allender, J. Bray, J. Bardeen, Phys. Rev. B **7**, 1020 (1973); Phys. Rev. B **8**, 4433 (1973).