

## Investigation of the influence of a magnetic field on the chemical potential of electrons in superconducting and ferromagnetic thin films

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It is experimentally shown that measurements of the chemical-potential variation in a magnetic field could give valuable information about the magnetization of thin films. For superconducting single-crystal  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin films two kinds of behavior of  $\mu(H)$  were observed depending on the substrate used. For films deposited on polished  $\text{NdGaO}_3$  single-crystal substrates the change of  $\mu$  was mainly reversible. That means that no magnetic flux was trapped during a magnetic-field sweep. For films deposited on  $\text{MgO}$ ,  $\text{SrTiO}_3$ , and  $\text{LaAlO}_3$  single-crystal substrates  $\mu$  changed irreversibly. The estimated critical current density is  $j_c \sim 10^7 \text{ A/cm}^2$  at  $T=4.2 \text{ K}$  in both cases. Oscillations of  $\mu$  in films evaporated on cleaved  $\text{MgO}$  substrates were observed. The effect of a magnetic field on  $\mu$  in a ferromagnetic film, predicted 60 years ago, was confirmed.

### I. INTRODUCTION

The chemical potential  $\mu$  is an important thermodynamical characteristic of electrons in a metal. There were several theoretical and experimental papers concerned with the influence of a magnetic field on  $\mu$ . Attention was drawn<sup>1,2</sup> to the fact that the de Haas-van Alphen (DHVA) oscillations of the density of states at the Fermi level, under the condition of constant electron density  $n$ , should lead to oscillations of  $\mu$  with an amplitude

$$\mu^{\text{osc}} = -kI^{\text{osc}}B/n \quad (1)$$

where  $I^{\text{osc}}$  is the amplitude of the DHVA oscillations of magnetization,  $B$  is the magnetic induction, and  $k$  is a coefficient dependent on the band structure (in a free-electron model  $k = \frac{2}{3}$ ).

For superconductors, simple considerations<sup>3</sup> give

$$\mu(H) = \mu(0) + H^2/8\pi \quad (2)$$

for sufficiently small fields. A magnetic field should also change  $\mu$  in ferromagnetic metals. This case will be discussed later.

In our earlier work we have searched for oscillations of  $\mu$  in single crystals of beryllium<sup>4</sup> and bismuth<sup>5</sup> but without success. Also no influence of a magnetic field on  $\mu$  was observed in bulk samples of superconducting niobium and ferromagnetic dysprosium.

The absence of DHVA oscillations of  $\mu$  in bulk samples was attributed to a compensating effect of the magnetostriction: the oscillations of  $\mu$  associated with oscillations in the density of states are canceled by oscillations of  $\mu$  caused by magnetostrictive changes of the volume.<sup>4,5</sup> Indeed, according to Ref. 6, magnetostrictive oscillations of volume in a free-electron model take the form

$$\delta V/V = -(2/3)\beta I^{\text{osc}}B \quad (3)$$

where  $\beta$  is the compressibility of a metal. Since

$$\mu \propto V^{-2/3} \quad (4)$$

and  $\beta = 3/2n\mu$  then the magnetostrictive change of chemical potential

$$\delta\mu = (2/3)I^{\text{osc}}B/n$$

that is exactly (1) but with opposite sign.

The magnetostriction itself may be considered as a result of virtual changes in chemical potential. Let us imagine that a bulk sample of a metal is inserted in a nonuniform magnetic field. If  $\mu$  is dependent on  $B$  then the electron density should be nonuniform: electrons will move from a region with high  $\mu$  to ones with low  $\mu$ . But due to condition of electroneutrality, the density of ions will change, too: it should be higher in regions of low  $\mu$ . This gives a net magnetostriction. Diminishing the volume in the region with low  $\mu$  will result in an increase in the chemical potential there [see Eq. (4)] until  $\mu$  becomes constant over the whole sample. Thus the chemical potential of electrons in bulk sample will not depend on a magnetic field. This constancy of  $\mu$  is simply one of the conditions of thermodynamical equilibrium.

On the other hand, large oscillations of  $\mu$  as a function of magnetic field were observed in the case of two-dimensional electron gas in heterojunctions<sup>5,7</sup> and in inversion layers.<sup>8</sup> This gave us the idea of eliminating the compensating effect of magnetostriction by using thin films instead of bulk samples because in this case the change of the film's volume is prevented by a substrate.

Let us consider unit volume of a metal film at a fixed temperature  $T$  in a varying magnetic field and apply the first law of thermodynamics, i.e., write down the equality between the change of internal energy of the electrons and the sum of thermal, mechanical, chemical and magnetic energies:

$$-IdB = TdS - pdV + nd\mu + BdI, \quad (5)$$

where  $S$  is the electronic entropy and  $p$  is the applied pressure. At low temperatures and neglecting changes in volume we obtain

$$n\delta\mu \approx -\delta(IB). \quad (6)$$

## II. EXPERIMENT

A natural way to detect the change of chemical potential in a magnetic field is to measure the contact potential difference between a test sample and a reference electrode, which constitutes two plates of a capacitor. This potential difference is equal to the difference in the work functions of the sample and the reference electrode

$$\Phi_r - \Phi_s = (-\mu - e\Delta\varphi)_r - (-\mu - e\Delta\varphi)_s, \quad (7)$$

where  $e$  is the electronic charge and  $\Delta\varphi = \varphi(\infty) - \varphi_i$  is a potential drop at the metal surface.<sup>9,10</sup> If the influence of a magnetic field on  $\Phi_r$  and  $\Delta\varphi_s$  is negligibly small then change in charge on the capacitor can be written as

$$\delta Q = C\delta\mu_s/e. \quad (8)$$

Using this method it is, of course, impossible to obtain absolute values of chemical potential, only variations from the mean value may be detected. An advantage of this method is the locality, because the mean value of  $\mu$  only arises from that part of the sample which is placed opposite to the reference electrode and of a thickness, which is of the order of the electric-field penetration depth, is measured.

A schematic diagram of our device is shown in Fig. 1. The measuring capacitor is mounted at the end of the coaxial line which is in turn connected to the electrometer. To reduce the noise, the coaxial line with measuring capacitor are inserted in a stainless-steel tube filled with gaseous helium ( $p \approx 200$  mm Hg at  $T = 300$  K). Care needs to be taken in constructing the coaxial line and the measuring capacitor. Our coaxial line consists of a stainless-steel tube and a copper wire insulated with sapphire insulators. Due to large piezoelectric or triboelectric effects it is impossible to use materials with high dielectric constant (like SrTiO<sub>3</sub> or Mylar) as insulators in the measuring capacitor. Thin cigarette or condenser paper works reasonably well at liquid-helium temperatures ( $C \approx 100$  pF/cm<sup>2</sup>), but it is better not to use a solid insulator at all. In the construction of the measuring capacitor, shown in Fig. 1, the reference electrode was insulated

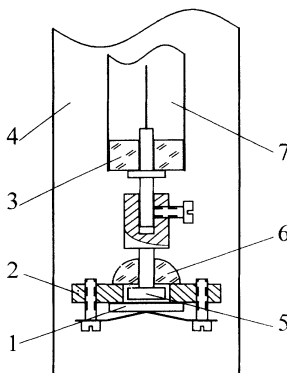


FIG. 1. Schematic diagram of the device for the chemical-potential measurements. 1—sample, 2—base plate, 3—sapphire insulator, 4—protective tube, 5—reference electrode, 6—glass insulator, 7—coaxial line to an electrometer.

from the base plate with glass. The reference electrode and base plate were made from bronze. A small slit ( $\approx 0.01$  mm) between the electrode and the sample pressed to the base plate arose after cooling the capacitor, due to the difference in thermal expansion between glass and bronze. For the electrode of 5 mm diameter the capacitance  $C \approx 10$  pF. The electrometer should have high sensitivity, low noise, and drift. We have used the electrometer with a vibrating capacitor at its input. This capacitor converted the charge on it to an alternative voltage ( $\approx 240$  Hz), which was amplified and lock-in detected. Calibration of the device was performed by connecting the sample to a known electric potential and measuring the induced charge.

Besides chemical potential, we have also measured the magnetic moment of some superconducting films. For these measurements the Hall probe and capacitance magnetometer based on the Faraday method were used. The idea of the capacitance magnetometer was taken from the work,<sup>11</sup> but instead of a capacitance bridge we have used an LC-generator with a metal-oxide-semiconductor field-effect transistor mounted near to the capacitance sensor. An oscillating circuit consisted of a small coil with  $L \approx 10$   $\mu$ H and capacitance sensor with  $C \approx 10$  pF. The sample was glued to the movable plate of the capacitance sensor and placed some 15–20 mm above the center of the magnet. The force acting on the film was calculated from the change in frequency of the LC generator.

Single-crystal films of high- $T_c$  superconductor YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$</sub>  (YBCO) used in this study were prepared by laser ablation.<sup>12</sup> Recently cleaved faces of MgO single crystal and polished plates of MgO, SrTiO<sub>3</sub>, LaAlO<sub>3</sub>, and NdGaO<sub>3</sub> single crystals were used as substrates. The dimensions were of the substrates  $1 \times (1-1.5)$  cm<sup>2</sup>. The crystallographic  $c$  axis was perpendicular to the film plane. Along one side of the film narrow gold strips for electrical contacts were evaporated.

The measurements were carried out at the International Laboratory of High Magnetic Field and Low Temperatures (Wroclaw, Poland) and at the Kapitza Institute for Physical Problems (Moscow, Russia). The magnetic field was produced by superconducting solenoids and a Bitter-type magnet.

## III. RESULTS FOR OSCILLATIONS OF THE CHEMICAL POTENTIAL

Oscillations of the chemical potential were observed for  $H \parallel c$  in single-crystal YBCO films with a thickness about 600 Å deposited on cleaved faces of the MgO. To separate the oscillations, at first a monotonic part fitted by parabola was subtracted from the recorded signal and then an average was taken over several ( $\sim 10$ ) measurements.

We have repeated measurements of the film, data for which were published in Ref. 12. In Fig. 2 old data are compared with the new data which were obtained half a year later. It is seen that a significant change has occurred in the spectrum of oscillations. If we suppose that observed oscillations are of the DHVA type, i.e., their period is constant in reciprocal magnetic field, then for

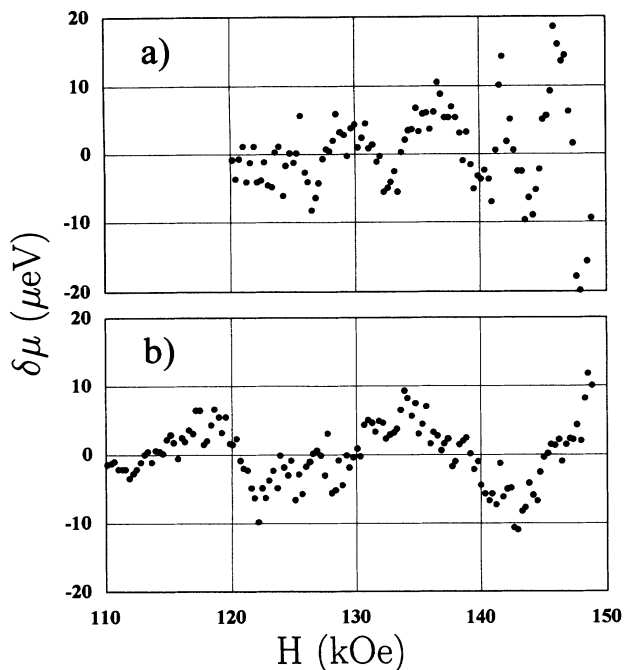


FIG. 2. Chemical-potential oscillations observed in YBCO film at  $T=1.4$  K: (a) data from 24.11.91, (b) data from 13.05.92.

the old data the oscillations with frequency  $F \approx 2.6$  MG dominate whereas for the new data the main frequency is  $F \approx 0.9$  MG. Such a large decrease in the frequency correlates with a diminishing  $T_c$  of the film. At the beginning  $T_c$  was equal to 85 K and it was lowered to  $T_c = 60$  K after a long storage.

Two recently prepared films were also measured. For

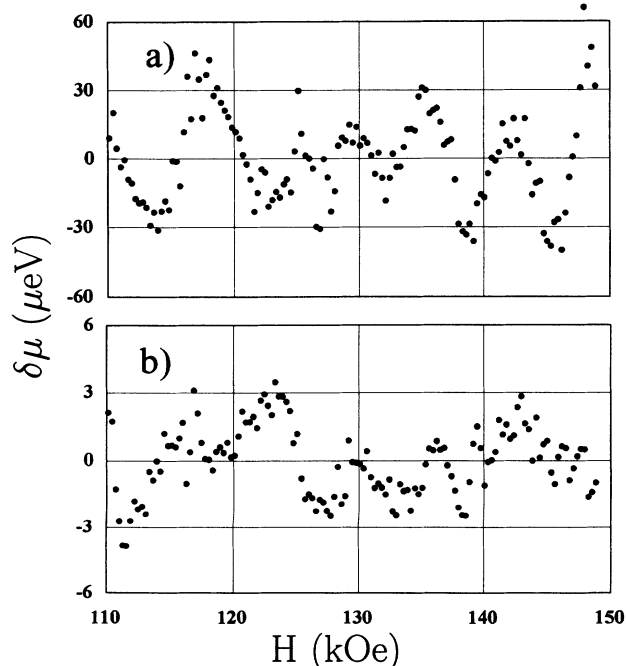


FIG. 3. Chemical-potential oscillations observed in recently prepared YBCO film at  $T=1.4$  K (a) and  $T=2.8$  K (b).

both films at  $T=1.4$  K oscillations of the chemical potential were observed with frequency  $F \approx 2.5$  MG for one film and  $F \approx 3$  MG for the other. Increasing the temperature resulted in a diminishing amplitude of the oscillations. For the data shown in Fig. 3 the amplitude of the main harmonic was  $12 \mu\text{eV}$  at  $T=1.4$  K and was about  $1.1 \mu\text{eV}$  at  $T=2.8$  K. The effective mass of the carriers, estimated from the amplitude ratio, is  $m^* \approx 2m$ .

#### IV. RESULTS FOR FILMS DEPOSITED ON POLISHED SUBSTRATES

In measurements of single-crystal YBCO films with thickness 500–3000 Å deposited on polished substrates, a large variation in the chemical potential of electrons was observed in a magnetic field perpendicular to the substrate plane. The size of this change points to the role of

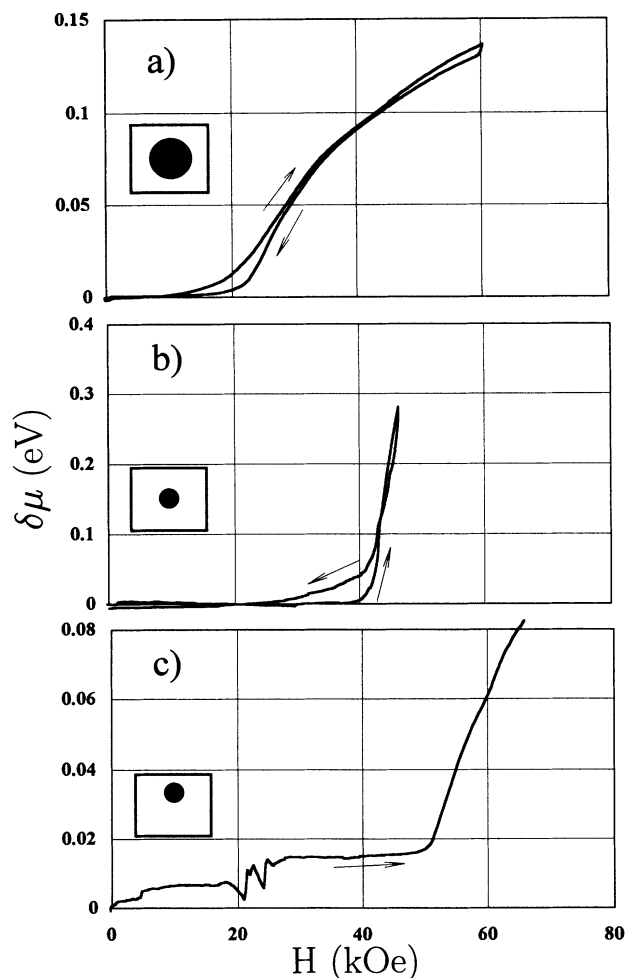


FIG. 4. The change in  $\mu$  of YBCO film with thickness 3000 Å deposited on polished  $\text{NdGaO}_3$  substrate and measured at  $T=4.2$  K in cases: (a) the reference electrode with radius  $a_r=2.5$  mm was placed in the center of the film, (b) the reference electrode with radius  $a_r=1$  mm was placed in the center of the film, (c) the reference electrode with radius  $a_r=1$  mm was shifted from the central position.

superconducting shielding currents. Two different types of behavior in  $\delta\mu(H)$  were observed: reversible and irreversible.

For films recently deposited on NdGaO<sub>3</sub> substrates, the chemical potential of the central part of the film did not change with increasing field up to some threshold value  $H_t$  (dependent on the radius  $a_r$  of the reference electrode), and then rapidly increased at higher field. In Fig. 4 several measurements for film with thickness 3000 Å are displayed. Measurement (a) was done for  $a_r=2.5$  mm (threshold field  $H_t \sim 18$  kOe) and only small hysteresis was observed. For measurement (b)  $a_r=1$  mm and  $H_t$  rose to 40 kOe. If the field was increased to values much higher than  $H_t$  then jumps in  $\mu$  were sometimes observed. The behavior of  $\mu$  was also dependent on the position of the reference electrode [Fig. 4(c)], and when it was placed above the corner of the film very large jumps in chemical potential were observed for some films.

The change of  $\mu$  was sensitive to the film history and to the way in which the grounding wire was connected to the gold strip. When a pressed indium contact was used the value  $H_t$  and the change of  $\mu$  were large. Substitution of pressed contacts for the soldered one resulted in decreasing  $H_t$  and  $\delta\mu$  (Fig. 5). This is probably a result of deterioration of the current capability of the film.

Some measurements were made using a large reference electrode isolated from the film by thin condenser paper, and the change of  $\mu$  of almost the whole film was detected. In this case  $\mu$  changed reversibly in fields up to 60–70 kOe [Fig. 6(a)]. In fields greater than 100 kOe the relaxation of  $\delta\mu$  with time became evident and a dimin-

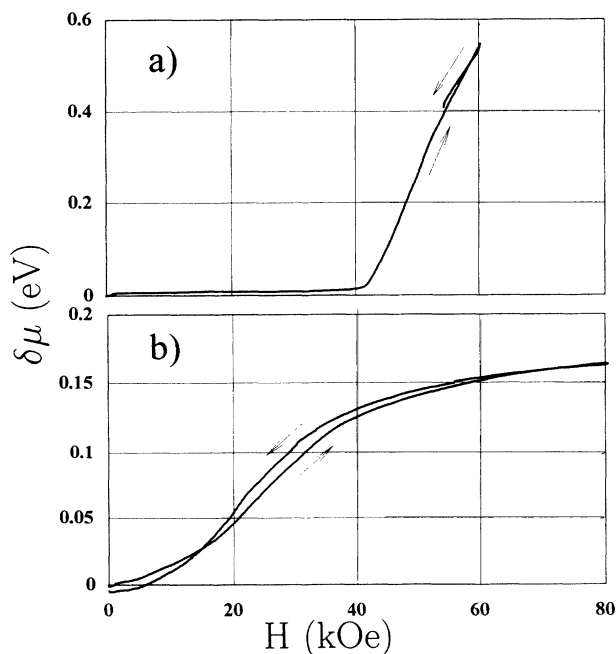


FIG. 5. The change in  $\mu$  of YBCO film with thickness 2000 Å deposited on polished NdGaO<sub>3</sub> substrate and measured at  $T=4.2$  K with the reference electrode with radius  $a_r=2.5$  mm when the grounding wire was pressed with indium (a) and after it was soldered (b).

ishing  $\delta\mu$  at  $H=150$  kOe [Fig. 6(a)] was seen due to this effect (the field was kept constant for two minutes). Lowering the temperature down to 1.45 K [Fig. 6(b)] had no influence on the relaxation rate.

Similar behavior was observed for a film with thickness 500 Å. In this case, the change of  $\mu$  in perpendicular magnetic field 150 kOe at  $T=4.2$  K exceeded 2.5 eV, that is two times larger than for the film with a thickness 2000 Å. This result may be connected with a less pronounced effect of the magnetostriction in the thinner film.

The relaxation of  $\delta\mu$  was dependent on the size of the reference electrode. From Fig. 7 it is seen that  $\delta\mu$  of the whole film noticeably relaxed at  $H=120$  kOe (b) whereas  $\delta\mu$  of the central part of the film did not change with time even at  $H=150$  kOe (a). This means that the relaxation is connected with the film edges.

For this film  $\delta\mu$  was measured at  $T=77$  K in the Bitter-type magnet. The change of  $\mu$  in this case was two orders smaller than at  $T=4.2$  K. Orientation of the field had a great influence on  $\delta\mu$ , too. The change of  $\mu$  with a field decreased by more than an order of magnitude in a parallel magnetic field [Fig. 6(c)].

It should be stressed that for films deposited on pol-

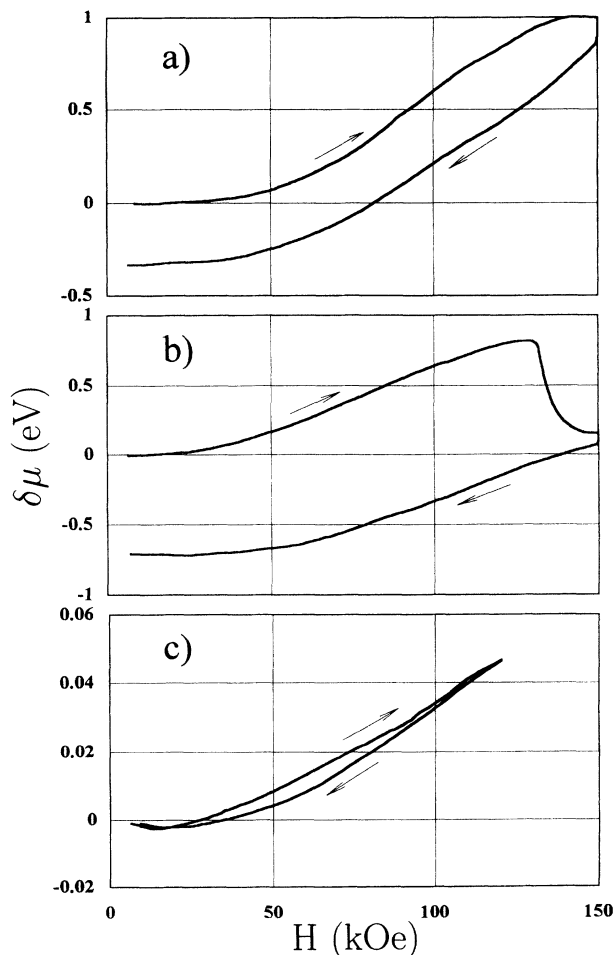


FIG. 6. The change in  $\mu$  of the whole YBCO film with thickness 2000 Å deposited on polished NdGaO<sub>3</sub> substrate and measured in cases: (a)  $H \parallel c$ ,  $T=4.2$  K; (b)  $H \parallel c$ ,  $T=1.45$  K; (c)  $H \perp c$ ,  $T=4.2$  K.

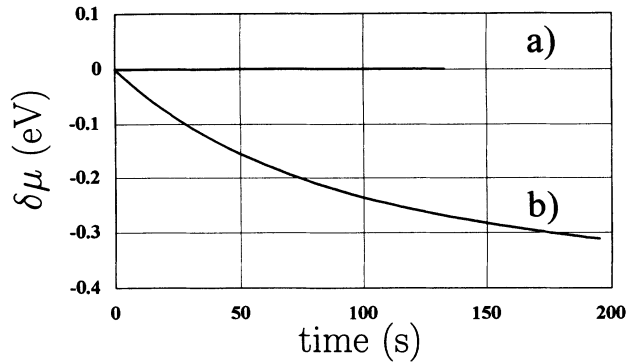


FIG. 7. Chemical-potential relaxation in YBCO film with thickness 500 Å deposited on polished NdGaO<sub>3</sub> substrate and measured at  $T=4.2$  K in cases: (a) the reference electrode with radius  $a_r=2.5$  mm was used,  $H=150$  kOe; (b) a large reference electrode was used,  $H=120$  kOe.

ished NdGaO<sub>3</sub> substrates the change of chemical potential was approximately reversible in spite of different dependences of  $\delta\mu$  on a magnetic field.

For films deposited on polished MgO, SrTiO<sub>3</sub>, and LaAlO<sub>3</sub> substrates the change in chemical potential was irreversible for fields higher than 0.5–1 kOe and we obtained hysteresis curved  $\delta\mu(H)$ . In Fig. 8 the measurement of  $\delta\mu$  with hysteresis loops obtained for film with thickness 3000 Å deposited on a polished LaAlO<sub>3</sub> substrate is depicted. In this case  $\delta\mu(H)$  was only slightly dependent on the radius of the reference electrode and its position over the film. In Fig. 9 the magnetic moment of the film measured by the capacitance magnetometer (a) and the product  $-MH$  (b) are shown. From a comparison of Fig. 8 and Fig. 9(b) it is seen that  $\delta\mu$  behaves similarly to  $-MH$  in agreement with Eq. (6).

We also performed an experiment with bulk polycrystalline sample YBCO. No variation in the chemical potential in a magnetic field was detected.

## V. RESULT FOR NICKEL

A nickel film with thickness 500 Å was prepared by laser vacuum deposition and measured in a parallel field

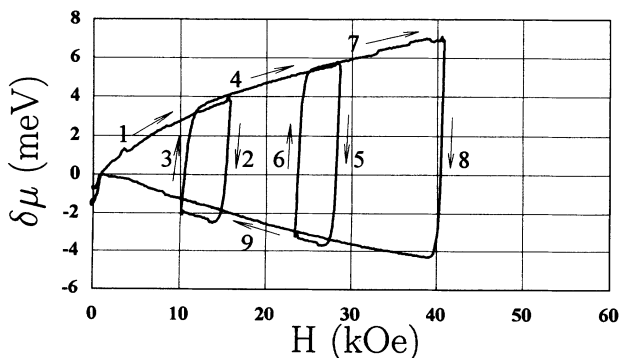


FIG. 8. The change in  $\mu$  of YBCO film with thickness 3000 Å deposited on polished LaAlO<sub>3</sub> substrate and measured at  $T=4.2$  K with the reference electrode with radius  $a_r=2.5$  mm.

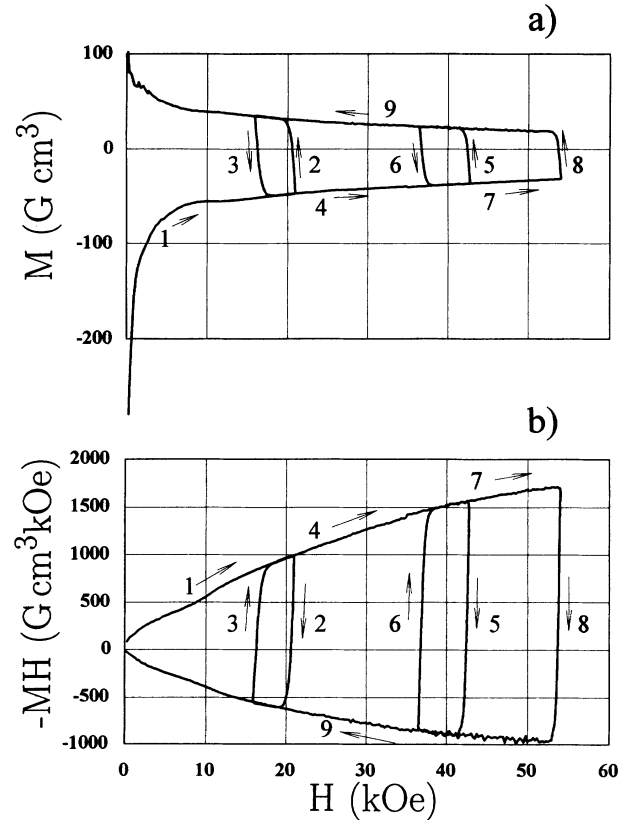


FIG. 9. (a) Magnetic moment of YBCO film with thickness 3000 Å deposited on polished LaAlO<sub>3</sub> substrate and measured at  $T=4.2$  K by the capacitance magnetometer. (b) The dependence of the product  $-MH$  on magnetic field.

at  $T=4.2$  K. Only one run up and down was performed. The result is detected in Fig. 10. A linear fit shown by the dashed line corresponds to  $\delta\mu[\mu\text{eV}]=4.56H[\text{kOe}]$ .

## VI. DISCUSSION

### A. Oscillations of chemical potential

Two different interpretations of the observed oscillations are possible. As in a short note<sup>12</sup> we may suppose

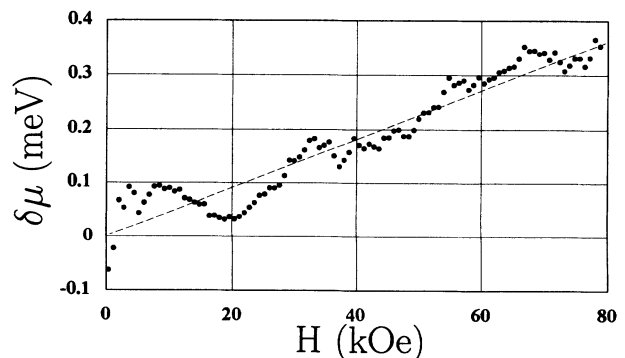


FIG. 10. The change in chemical potential of nickel film with thickness 500 Å at  $T=4.2$  K in parallel magnetic field. The dashed line shows linear fit.

that they result from the DHVA effect. Up to now two experiments have been performed in which the DHVA effect was detected on oriented powders of YBCO embedded in epoxy.<sup>13-16</sup> In pulsed magnetic fields 650–1000 kOe, oscillations with frequencies  $F=5.3, 7.8,$  and  $35.1$  MG and effective masses  $m^* \approx 7m$  were observed.<sup>13-15</sup> In stationary magnetic fields 135–240 kOe oscillations with frequency  $F \approx 5.4$  MG and  $m^* \approx 2.1m$  were observed.<sup>16</sup> In our data for recently prepared YBCO films, oscillations with frequency  $F=2.5-3$  MG and  $m^* \approx 2m$  dominate. Fourier spectrum shown in Fig. 2 of our old work<sup>12</sup> contains a component with  $F \approx 5.2$  MG but this is probably the second harmonic of the main frequency. The observed oscillations of  $\mu$  may be ascribed to the hole tubes of the Fermi surface surrounding  $S$ - $R$  edges of the Brillouin zone, when compared with calculations.<sup>17,18</sup> At first sight a diminishing frequency  $F$  after prolonged storage (Fig. 2) confirms the DHVA origin of oscillations of  $\mu$ . Simultaneous lowering of the critical temperature implies a decreasing carrier concentration which should result in diminishing Fermi-surface dimensions. However, the measured critical temperature ( $T_c \approx 60$  K) corresponds to an oxygen deficiency  $\delta \approx 0.4$  and according to Ref. 17 the hole tubes should disappear at  $\delta \approx 0.1$ . The most crucial peculiarity of the data is the constancy of the oscillation period seen in Fig. 3. For the DHVA effect the period should increase as  $H^2$ .

On the other hand, it is possible to explain the oscillations of  $\mu$  as a result of mesoscopic effects arising due to weak-link currents. In Sec. III we mentioned that oscillations were observed in a large field-dependent background. As this background is certainly connected with magnetization from the induced shielding currents, small oscillations of this magnetization could lead to oscillations of the chemical potential. An effective mass obtained in our measurements  $m^* \approx 2m$  also supports this point of view. From this period of oscillations  $\Delta H \approx 10$  kOe (Fig. 3) we can estimate mean loop size  $l = \sqrt{\Phi_0/\Delta H} \approx 400$  Å, where  $\Phi_0$  is the flux quantum. This value is in agreement with the dimensions of the steplike structure in the cleaved MgO substrate.<sup>19</sup> The increase of the period observed after prolonged storage [Fig. 2(b)] may be ascribed in this case to decreasing loop size due to the creation new weak links.

### B. Change of the chemical potential due to shielding currents

As was described in Sec. IV, we have observed two types of behavior in the chemical potential of films deposited on polished substrates: reversible and irreversible. For YBCO films deposited on polished NdGaO<sub>3</sub> substrates and not spoiled by soldering the ground wire, the chemical potential rapidly and reversibly increased above a threshold magnetic field  $H_t$ , dependent on the radius and position of the reference electrode. Below this value no variation of  $\mu$  was observed. This means that for  $H < H_t$  the magnetic induction in the central part of the film is  $B=0$  due to shielding by circulating currents. When the field increases the inner radius  $a_c$  of this current ring decreases and at  $H=H_t$  it becomes equal to the reference electrode radius  $a$ , [Figs. 4(a) and 4(b)]. At

some value of magnetic field  $a_c=0$  and current lines distort from circular. This results in a visible change of the  $\delta\mu(H)$  dependence seen in Fig. 4(c) at  $H \approx 52$  kOe. This picture leads to the dependence of the threshold field  $H_t$  on the inner radius of current ring shown in Fig. 11.

Let us compare our data with Eq. (6). In order to make some estimations of  $n$  and  $j_c$  we assume the following behavior for induction  $B$  and magnetization  $I$ :

$$B(r) = \begin{cases} 0, & r \leq a_c \\ \frac{r-a_c}{R-a_c} H, & r > a_c, \end{cases} \quad (9)$$

$$I(r) = \begin{cases} I_0, & r \leq a_c \\ \frac{R-r}{R-a_c} I_0, & r > a_c, \end{cases}$$

where  $R$  is the position of the film's edge. The value  $I_0$  may be obtained by assuming that the central part of the sample is in the Meissner state:<sup>20</sup>

$$I_0 = -\frac{2a_c}{3\pi^2 d} H, \quad (10)$$

where  $d$  is thickness of the film.

Using (6) we obtain for  $H - H_t \ll H_t$

$$nV\delta\mu = n\pi a_r^2 d \delta\mu = - \int BI dV \approx \frac{2a_c^2(a_r - a_c)^2}{3\pi(R - a_r)} H^2. \quad (11)$$

From Fig. 11 we have

$$(a_r - a_c) = R[H - H_t(a_r)]/H_t(a_r).$$

For the experimental data shown in Fig. 4(b),  $a_r=0.1$  cm,  $R=0.5$  cm,  $H_t=40.52$  kOe,  $\delta\mu/(H - H_t)^2 \approx 1.28 \times 10^{-8}$  eV/(Oe)<sup>2</sup>, and we obtain  $n \approx 1.2 \times 10^{23}$  cm<sup>-3</sup>. This value is comparable with the density of valence electrons in YBCO, which is equal to  $98/v_{\text{cell}} \approx 5.4 \times 10^{23}$  cm<sup>-3</sup>.

The dependence of  $H_t$  on  $a_c$  shown in Fig. 11 may be used to estimate the critical current density. For this purpose the equality between the magnetic moment of

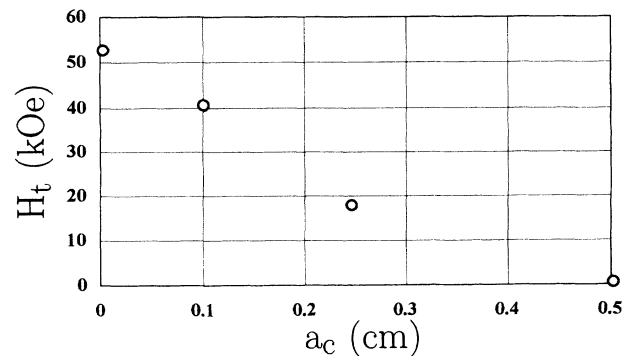


FIG. 11. The dependence of the threshold field on the inner radius of the current ring in YBCO film with thickness 3000 Å deposited on polished NdGaO<sub>3</sub> substrate.

the superconducting disk with radius  $a_c$  [Eq. (10)] and the magnetic moment of the current ring with inner radius  $a_c$  and outer radius  $R$  may be written. Assuming a constant value of  $j_c$  over the current ring we obtain

$$\frac{2a_c^3}{3\pi} H = \frac{\pi}{3c} j_c (R - a_c) (R^2 + Ra_c + a_c^2) d . \quad (12)$$

For  $H = 40$  kOe,  $j_c \approx 2.2 \times 10^7$  A/cm<sup>2</sup>.

When films are spoiled by soldering the grounding wire or by prolonged exposure to moist air then the threshold field disappears [Fig. 5(b)]. It may be connected with large variations in the critical current density over the films in this case. Nevertheless  $\delta\mu(H)$  curves remain reversible. This means that no magnetic flux is trapped during magnetic-field sweeps. Reversibility of the magnetic moment of a YBCO film with thickness 500 Å deposited on polished NdGaO<sub>3</sub> substrate was confirmed by direct measurements with the capacitance magnetometer. In fields up to 30 kOe the magnetic moment changed reversibly and was proportional to  $-H^{1.8}$ . As it is expected from Eq. (6) the chemical potential of the whole film measured in the same field range varied as  $\delta\mu \propto H^{2.8}$ .

For the films deposited on other substrates, the chemical potential changed irreversibly (Fig. 8), that is, the magnetic flux easily penetrated the film in increasing field and was trapped in decreasing field. Approximate constancy of  $\delta\mu$  over the film means that in this case the magnetic moment of induced currents was distributed uniformly with density

$$m = j_c R d / c , \quad (13)$$

where  $c$  is the velocity of light.

The uniformity of the magnetic moment was checked by measuring the magnetic field near the center of the film with a small Hall probe placed at distance  $z \approx 1$  mm under it.

Our film had rectangular shape with area  $S = 1 \times 1.5$  cm<sup>2</sup>. For simplicity of calculation let us replace this by a disk with radius  $R = \sqrt{S/\pi}$ . If the magnetic moment is distributed uniformly, then readings of the Hall probe will differ from the applied magnetic field by an amount

$$\Delta H = \frac{2\pi R^2}{(R^2 + z^2)^{3/2}} m \quad (14)$$

and the whole moment of the film

$$M = \pi R^2 m , \quad (15)$$

where  $m$  is given by Eq. (13).

Putting in (14) and (15) the values of  $R$  and  $z$  we obtain

$$\Delta H [\text{Oe}] \approx 5.9 M [\text{G cm}^3] . \quad (16)$$

But if the current ring is formed, then

$$\Delta H = \frac{2\pi}{R} \left[ \ln \left| \frac{R + \sqrt{R^2 + z^2}}{z} \right| - \frac{R}{\sqrt{R^2 + z^2}} \right] \frac{J}{c} , \quad (17)$$

$$M = \frac{\pi}{3} R^2 \frac{J}{c} , \quad (18)$$

where  $J$  is the whole current through the ring with inner

radius  $a = 0$ , and for our case

$$\Delta H [\text{Oe}] \approx 30 M [\text{G cm}^3] . \quad (19)$$

Experimental results for  $H = 20$  kOe are

$$M = -48 \text{ G cm}^3 , \quad \Delta H = -250 \text{ Oe} .$$

This is in good agreement with (16) but not with (19).

It is interesting to note that the critical current density estimated for this film with Eq. (13),

$$j_c (20 \text{ kOe}) \approx 1.6 \times 10^7 \text{ A/cm}^2 ,$$

$$j_c (40 \text{ kOe}) \approx 1.2 \times 10^7 \text{ A/cm}^2 ,$$

is the same order of magnitude as for the YBCO film deposited on a NdGaO<sub>3</sub> substrate.

Thus the difference in their behavior is not associated with the value of critical current itself. It should be stressed that usually our films were deposited on different substrates by the same process at the same time, so technological reasons are excluded. Two circumstances may be pointed out. (i) Unlike other materials used for substrates, the NdGaO<sub>3</sub> lattice ideally matches YBCO (Ref. 21). (ii) It has a localized magnetic moment of Nd<sup>3+</sup> with  $\mu_{\text{eff}} = 3\mu_B$ .

The first reason seems to be more important. According to Ref. 21 the lattice mismatch of NdGaO<sub>3</sub> with YBCO is 0.2% whereas for other substrates the mismatch is about 2%. As a result, the films with thickness larger than 150 Å deposited on SrTiO<sub>3</sub>, LaAlO<sub>3</sub>, and MgO substrates are not flat and smooth but have a mosaic texture. In this case magnetic flux easily penetrates between the grains of the film even at low applied external magnetic fields due to the locally enhanced stray field.<sup>22</sup> In contrast, the films deposited on the NdGaO<sub>3</sub> substrate have no mosaic texture.<sup>21</sup>

### C. Change of the chemical potential in nickel film

The problem of influence of a magnetic field on the chemical potential of electrons in ferromagnetic metals was formulated by Dorfman,<sup>23</sup> but an attempt to observe this effect was unsuccessful.<sup>24</sup> Formulation in modern terms was given by Walmsley<sup>25</sup> 30 years later.

In the ferromagnetic state bands of  $d$  electrons with spin-up ( $d^+$ ) and spin-down ( $d^-$ ) are shifted in energy due to the exchange interaction. If we apply an external field, then the energy of  $d^+$  electrons decreases and  $d^-$  electrons increases by  $\mu_{\text{eff}} H$ . The resulting change of the chemical potential may be found from the condition of constancy of the electron concentration

$$(\delta\mu + \mu_{\text{eff}} H) g^+ + (\delta\mu - \mu_{\text{eff}} H) g^- = 0 , \quad (20)$$

where  $g$  denotes density of states at the Fermi level. Thus

$$\delta\mu = \frac{g^- - g^+}{g^- + g^+} \mu_{\text{eff}} H . \quad (21)$$

In nickel the Fermi level lies just above  $d^+$  band<sup>26</sup> so  $g^+ = 0$  and we get

$$\delta\mu^{(J)} = \mu_{\text{eff}} H . \quad (22)$$

This is the first effect of a magnetic field on the chemical potential of electrons in a ferromagnetic film. The second effect is associated with its magnetization and is described by Eq. (6):

$$N_v \delta\mu^{(II)} = -\mu_{\text{eff}} H, \quad (23)$$

where  $N_v$  is the number of valence electrons per atom.

In the case of nickel  $N_v = 10$  and the resulting change in the chemical potential

$$\delta\mu = \delta\mu^{(I)} + \delta\mu^{(II)} = (1 - 1/N_v)\mu_{\text{eff}} H \approx 0.9\mu_{\text{eff}} H. \quad (24)$$

For the results shown in Fig. 10,  $\mu_{\text{eff}} \approx 0.88\mu_B$ . This value is about 1.5 times larger than any value reported in the literature,<sup>27</sup> but it should be noted that published data were obtained at constant pressure whereas our data were obtained at constant volume.

Previous attempts to observe this effect<sup>25,28</sup> were unsuccessful, mainly due to the choice of dielectric for the measuring capacitor. For example, in Ref. 28, SrTiO<sub>3</sub> was used and magnetostriction of the ferromagnetic film resulted in a large piezoelectric signal. In our experiment

we did not use solid dielectric at all, but it was necessary to have a high sensitivity electrometer.

## VII. CONCLUSION

In this paper we have shown the applicability of the chemical-potential measurements for the investigation of the magnetic properties of thin films. For superconducting films this method permits us to obtain the local distribution of magnetization with a resolution equal to the size of reference electrode. In this sense it is similar to magneto-optical methods but may be used in much higher magnetic fields. Owing to the resolution in depth this method may be especially useful in the case of superconducting multilayers and magnetic superlattices.

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