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# Temperature dependences of the surface resistance and the diamagnetic shielding susceptibility at $T_c - T \ll T_c$ for high- $T_c$ superconductors\*\*

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At  $T_c - T \ll T_c$  (i.e., near  $T_c$ ), in order to demonstrate the conduction mechanism and temperature dependencies of the diamagnetic-shielding susceptibility and the penetration depth, we fabricated  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$  (BKBO) thin films and measured the energy gap by tunnel effect and shielding susceptibilities which are compared with those measured for BKBO and YBCO single crystals. The shielding susceptibilities for BKBO and YBCO crystals well-fit  $\chi(T)/\chi(0) = 1 - \exp(-2\Delta(T)/k_B T)$ , while that for the BKBO film follows  $\chi(T)/\chi(0) = (1 - T/T_c)$  which may not be intrinsic. The exponential decrease of the susceptibilities near  $T_c$  indicates that the conduction mechanism is hopping. The energy gaps are observed as  $2\Delta(0) = (3.5 \pm 0.1)k_B T_c$  for the BKBO film by the tunnel effect,  $2\Delta(0) = (3.9 \pm 0.1)k_B T_c$  for the BKBO single crystal, and  $2\Delta(0) = (8 \pm 0.2)k_B T_c$  for the  $\text{YBa}_2\text{Cu}_3\text{O}_7$  single crystal. Furthermore, for microwave device applications of superconductors, at  $T_c - T \ll T_c$ , the surface resistance  $R_s(T) \approx \sqrt{\frac{\omega\mu_0/2}{\sigma_n + (\sigma_s(0) - \sigma_n)f(T)}}$  is derived from the surface impedance at  $\omega\tau_{tr} \ll 1$ , where  $\sigma_s(0)$  and  $\sigma_n$  are the conductivities of the superconducting state and the normal state, respectively, and  $f(T) = \chi(T)/\chi(0) = (1 - \exp(-2\Delta(T)/k_B T))$ .

## I. INTRODUCTION

The shielding susceptibility,  $\chi$ , was suggested as a means of observing the London penetration depth,  $\lambda_L$ .<sup>1-3</sup>  $\lambda_L$  has been known as a direct measure of the superfluid density and a probe of the pairing state of superconductors. For experimental results for high- $T_c$  superconductors at  $T \ll T_c$ ,  $\lambda_L(T)$  has exhibited a  $T^2$  temperature dependence<sup>4</sup>, as well as a linear  $T$  dependence<sup>5</sup>. The linear  $T$  dependence has been predicted in  $d$ -wave superconductivity. Recently,  $\lambda_L(T)$ , which is independent of temperature, was demonstrated.<sup>3</sup>

On the other hand, for the 3 dimensional(D) XY behavior which enhances fluctuations in the order parameter  $\Psi$  for Ginzburg-Landau (GL) theory, the diamagnetic shielding susceptibility was given by  $\chi(T) = -(kT/\Phi_0^2)(T - T_c)^{-0.67}$ , while the GL result  $\chi(T) \approx (T - T_{co})^{-0.5}$  near  $T_c$ .<sup>6</sup> The GL result was suggested to need modification [6]. The 3D XY behavior was absent in the magnetic penetration depth of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) films.<sup>7</sup> Moreover, at  $T_c - T \ll T_c$ , from the BCS picture  $\chi(T)/\chi(0) = (1 - T/T_c)$  was derived and from the phenomenological model  $\chi(T)/\chi(0) = 1 - \exp(-\frac{2\Delta(T)}{k_B T})$  was obtained.<sup>3,8,9</sup>

Furthermore, since 1960's, it has been known that BCS calculations deviate from experimental data of the electronic specific-heat contribution  $C_{es}$  for superconductive lead above the condensation temperature [10]. This suggests that a conduction mechanism instead of tunneling one needs at  $T_c - T \ll T_c$ .

For microwave device applications of superconductors, the measurement and the physical analysis of the surface impedance are very important.<sup>11</sup> In particular, its temperature dependence is still controversial at  $T_c - T \ll T_c$ ,<sup>12</sup> since the conduction mechanism with respect to

conduction carriers has not been clearly understood at  $T_c - T \ll T_c$ .

In this paper, we demonstrate experimentally the unclear temperature dependencies of  $\chi(T)$  and  $\lambda_L(T)$  at  $T_c - T \ll T_c$  for shielding susceptibilities measured for a BKBO film, high quality  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$  (BKBO) and YBCO crystals. The energy gap of the BKBO film is obtained from the tunnel effect, while energy gaps of BKBO and YBCO crystals are obtained from the fitting parameter of shielding susceptibilities. For microwave device applications of superconductors, the temperature dependence of the surface resistance is derived.

## II. THEORETICAL CONSIDERATION

The superconducting state as Meissner state is a condensed state. With increasing temperature, Cooper pairs surpass the superconducting energy gap to the excited state by the excitation of thermal phonons; this means pair-breaking which characterizes a superconductor-metal transition. In assuming that the diamagnetic-shielding susceptibility is proportional to the superfluid density in the superconducting state ( $\chi \propto n_{sup}$ ), in the microscopic theory,<sup>3,8</sup> the susceptibility was given by,

at  $T \ll T_c$ ,

$$\chi(T)/\chi(0) \approx 1 - \sqrt{(2\pi\Delta(0)/k_B T)} e^{-\Delta(0)/k_B T}, \quad (1)$$

and, at  $T_c - T \ll T_c$ ,

$$\chi(T)/\chi(0) = (1 - T/T_c). \quad (2)$$

Furthermore, at  $T_c - T \ll T_c$ , another susceptibility was given by

$$\chi(T) = \chi(0)(1 - \exp(-\frac{2\Delta(T)}{k_B T})). \quad (3)$$

Equation (3) was obtained from the simple phenomenological model based on the classical limit of Fermi-Dirac(FD) statistics (*i.e.*, Maxwell-Boltzman statistics) because the system is far from condensation; in the system the fluid density is very small and temperature is high relatively to  $T_c$ . Eq. (3) is the hopping one of the conduction mechanism for semiconductors and was given elsewhere.<sup>3,9</sup>

The susceptibility as a function of the penetration depth has been defined by

$$\chi(T)/\chi(0) = (\lambda_L(0)/\lambda_L(T))^2. \quad (4)$$

In addition, Eq (1) was demonstrated in a previous paper<sup>3</sup>.

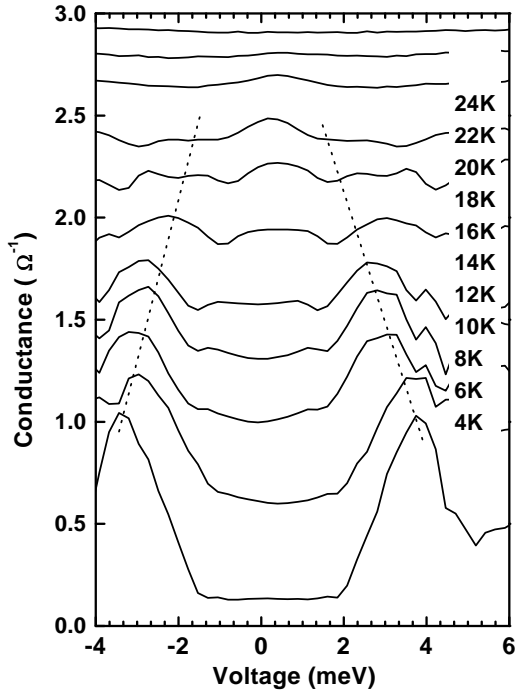


FIG. 1. Temperature dependence of the conductance measured by the tunnel effect for the BKBO25 thin film; from 4 to 24 K.

### III. EXPERIMENT

BKBO thin films were fabricated on (100)SrTiO<sub>3</sub> substrates in the argon atmosphere of 1 Torr at the substrate temperature of 520° by laser ablation. The deposition method and conditions were shown in a previous

paper<sup>13,14</sup>. The magnetic shielding susceptibilities corresponding to the zero field cooled (ZFC) susceptibility were measured at 10 Oe//c-axis by using Quantum Design SQUID. The superconducting magnet of the SQUID was quenched by perfect evaporation of liquid helium before the measurement. Moreover the ZFC susceptibilities for BKBO and YBCO single crystals were measured by the same method as that for the BKBO film.

In order to measure the temperature dependence of the superconducting energy gap for BKBO films, the conductances were measured by the point-contact method through the metal-superconductor-metal junction. The measurement temperatures were from 4.2 K to 24 K.

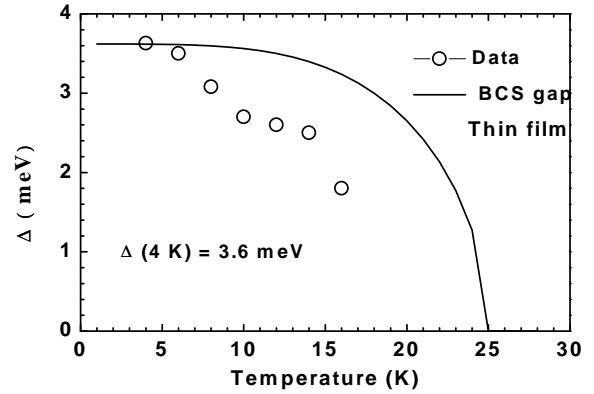


FIG. 2. Comparison of the energy gap measured for the BKBO25 film with the empirical energy gap of the BCS one,  $\Delta(T)/\Delta(0) \approx \sqrt{\cos[(\pi/2)(T/T_c)^2]}$  and  $2\Delta(0) = bk_B T_c$ . Here  $b \approx 3.5$ .

### IV. RESULTS AND DISCUSSION

In order to check the validity of the linear T dependence in Eq. (2), the temperature dependence of the conductance (or energy gap), measured by the tunnel effect for a BKBO thin film (named BKBO25) with  $T_c \approx 25$  K, is shown in Fig. 1. The conductance at 4 K has a clean U shape which is known as the energy gap with  $\Delta(4 \text{ K}) \approx 3.6$  meV. U curve begins to be deformed at 6 K, is observed up to 12 K, and is not seen from 16 to 24 K. This indicates that the energy gap forms below 14 K. Fig. 2 shows the comparison of the measured energy gap with the BCS gap with  $b \approx 3.5$ . The measured gap deviates from the BCS gap with increasing temperature. The deviation may arise from the pinning effect or vortices due to impurity phases in the film. Fig. 3 shows temperature dependencies of the susceptibility obtained from both Eq. (3) and  $\Delta(T)$  determined from the tunnel effect in Fig. 2, (called the "tunnel susceptibility"), the susceptibility obtained from both Eq. (3) and the BCS gap, (called the "BCS susceptibility"), and the ZFC susceptibility data measured for the BKBO25 film at 10 Oe. Below 14 K, the normalized susceptibilities agree closely.

The figure shows that the ZFC susceptibility data with linear  $T$  dependence at  $T_c - T \ll T_c$  follow the tunnel susceptibility and deviate from the BCS susceptibility. This indicates that crystals or films with an energy gap which does not follow the BCS gap at  $T_c - T \ll T_c$  exhibit a linear  $T$  dependence in the susceptibility, and that Eq. (2) is not intrinsic.

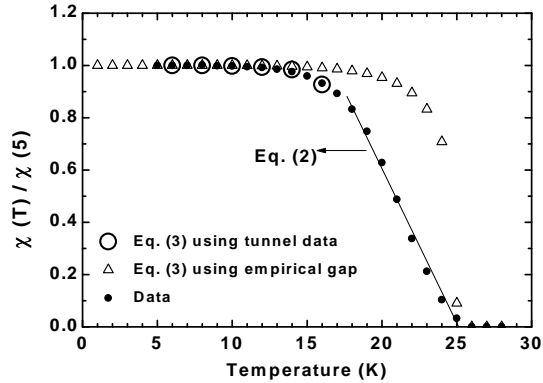


FIG. 3. Temperature dependencies of the susceptibility ( $\circ$ ) obtained from Eq. (3) using the energy gap determined from the tunnel effect in Fig. 1; the susceptibility ( $\Delta$ ) obtained from Eq. (3) using the empirical gap; and the ZFC susceptibility data ( $\bullet$ ) measured directly at 10 Oe for the BKBO25 film. At  $T_c - T \ll T_c$ , the linear is Eq. (2).

At  $T_c - T \ll T_c$ , the susceptibility data measured for the BKBO single crystal are well fitted by Eq. (3) using the empirical gap ( $\Delta(T)/\Delta(0) \approx \sqrt{\cos[(\pi/2)(T/T_c)^2]}$ ) of the BCS gap, as shown by the thick line in Fig. 4. The energy gap is obtained from the fitting parameter ( $2\Delta(0) = bk_B T_c$ ) as well and is given as  $2\Delta(0) = (3.9 \pm 0.1)k_B T_c$ . The energy gap of the single crystal agrees well with  $2\Delta(0) \approx 3.8k_B T_c$  which is determined by the tunnel effect [15]. Here, although Eq. (3) is well-fitted from low temperatures to  $T_c$ , the fitting cannot be believed below a temperature ( $T \approx 17$  K) in which the energy gap deviate from the BCS gap, as shown in the inset of Fig. 4.

Figure 5 shows the susceptibility measured at 10 Oe// $c$ -axis for a YBCO single crystal like cube with the size of  $1.3 \times 1.1 \times 1$  mm<sup>3</sup> and  $T_c \approx 93.2$  K, grown by a melting method. Eq. (1) and Eq. (3) are well applied to the susceptibility data below 45 K and above 80 K, respectively, as shown in Fig. 5 and its inset. The susceptibility in the intermediate temperature range from 45 to 80 K is not explained by the combined Eqs. (1) and (3), which is different from the BKBO case. The 45 K is regarded as the condensation temperature.

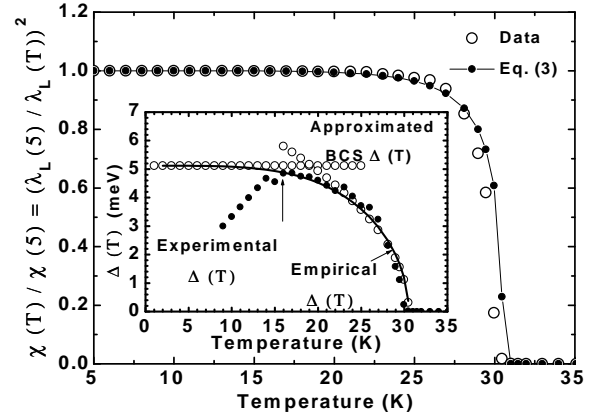


FIG. 4. The ZFC susceptibility measured at 10 Oe// $c$ -axis for the BKBO single crystal. The open circles ( $\circ$ ) are experimental data. The thick line and solid circles ( $\bullet$ ) indicates the fitting of Eq. (3) using the empirical energy gap. In the inset, the approximated BCS gap is  $\Delta(T) = 3.2k_B T_c \sqrt{1 - T/T_c}$  near  $T_c$  and  $2\Delta(0) = bk_B T_c$  at  $T=0$  K; the empirical gap is  $\Delta(T)/\Delta(0) \approx \sqrt{\cos[(\pi/2)(T/T_c)^2]}$ ; the experimental gap is deduced from Eq. (3) and is  $\Delta(T) = -(k_B T/2) \ln(1 - \chi(T)/\chi(0))$  where  $\chi(T)/\chi(0)$  uses the experimental susceptibility data. Here,  $b=3.9$ .

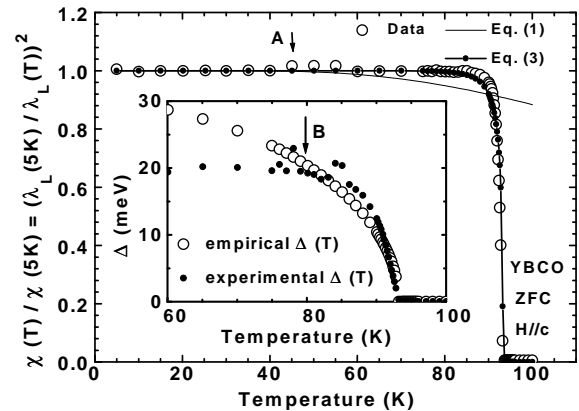


FIG. 5. Temperature dependence of the ZFC susceptibility measured at 10 Oe// $c$ -axis and its fitting for the YBCO single crystal with  $2\Delta(0) \approx 8k_B T_c$  and  $T_c \approx 93.2$  K. The susceptibility data ( $\circ$ ); Eq. (1) (thin line); and Eq. (3) (thick line and solid circles ( $\bullet$ )). The inset shows the empirical gap and the experimental one.

## V. SURFACE RESISTANCE

The temperature dependence of the surface resistance,  $R_s(T)$ , can be calculated by using the above results, which is necessary for microwave device applications of superconductors. Eq. (3) was derived

by assuming  $\chi(T)/\chi(0) = n_{sup}(T)/n_{tot} = f(T)$  and  $exp(-2\Delta(T)/k_B T) = n_{th}/n_{tot}$ ,<sup>3,8,9</sup> where  $n_{tot} = n_{sup} + n_{th}$ ,  $n_{tot}$  is the total number of pairs,  $n_{sup}$  denotes the number of Cooper pairs, and  $n_{th}$  stands for the number of thermally excited pairs (*i.e.*, carriers in the normal state). Eq. (1) was also given by  $\chi(T)/\chi(0) = n_{sup}(T)/n_{tot} = f(T)$ . The temperature dependence of  $\chi(T)$  corresponds to that of Cooper pairs. The above consideration is similar to the two fluid model.

From the London equation, the conductivity of the superconducting state is given by  $\sigma_s = \frac{(n_{sup}(\omega, T)/n_{tot})}{j\mu_0\omega\lambda^2(0)}$  at  $0 < \omega < \omega_s = 2\Delta(0)/\hbar$ , by using Eq. (4). Here,  $n_{sup}(\omega, T) \approx n_{sup}(T)$ , because Eq. (3) is valid at  $\omega \ll \omega_s$ . The temperature dependence of  $\sigma_s(T)$  is given by, at  $T \ll T_c$ ,

$$\sigma_s(0) = \frac{1}{j\mu_0\omega\lambda^2(0)}, \quad (5)$$

because of  $n_{th} \approx 0$  and  $f(T) = (n_{sup}(T)/n_{tot}) \approx 1$  in Eq. (1), while, at  $T_c - T \ll T_c$ ,

$$\sigma_s(T) = f(T)\sigma_s(0) = \frac{f(T)}{j\mu_0\omega\lambda^2(0)}, \quad (6)$$

where  $f(T) = 1 - exp(-2\Delta(T)/k_B T)$ . Moreover, because  $n_{th}$  is large at  $T_c - T \ll T_c$ , the normal conductivity,  $\sigma_n$ , occurring by  $n_{th}$  cannot be ignored. Thus the total conductivity is given by

$$\sigma_T(T) = \left(\frac{n_{sup}}{n_{tot}}\right)\sigma_s(0) + \left(\frac{n_{th}}{n_{tot}}\right)\sigma_n, \quad (7)$$

$$= f(T)\sigma_s(0) + (1 - f(T))\sigma_n, \quad (8)$$

where  $\sigma_n = \sigma_0/(1 + j\omega\tau_{tr})$  [12].

In order to obtain the surface impedance  $Z_s = \sqrt{j\omega\mu_0/\sigma_T}$  for high quality superconductors below the condensation temperature,  $T_s$ ,  $Z_s$  is important for microwave device application;  $T_s \approx 17\text{K}$  for BKBO and  $T_s \approx 80\text{K}$  for YBCO, as shown in Figs. (4) and (5). The total conductivity is approximated by  $\sigma_T \approx \sigma_s$ , since  $n_{th}$  can be ignored below  $T_s$  because it is very small. The surface impedance is given by, at  $T \ll T_c$ ,

$$Z_s = \sqrt{j\omega\mu_0/\sigma_s} \approx j\mu_0\omega\lambda(0), \quad (9)$$

while, at  $T_c - T \ll T_c$ ,

$$Z_s \approx \sqrt{j\omega\mu_0/(f(T)\sigma_s(0) + (1 - f(T))\sigma_n)}, \quad (10)$$

by using Eq. 8. At  $T \ll T_c$ ,  $Z_s$  is called the surface reactance  $X_s$ . At  $T_c - T \ll T_c$ ,  $Z_s$  can be expressed as  $Z_s = R_s + jX_s$  and the surface resistance is given by

$$R_s(T) \approx \sqrt{\frac{\omega\mu_0/2}{\sigma_n + (\sigma_s(0) - \sigma_n)f(T)}}. \quad (11)$$

This  $R_s$  is obtained by calculations of Eq. 10 at  $\omega\tau_{tr} \ll 1$ , increases with increasing temperature up to  $T_c$ , and depends upon  $n_{sup}(\propto f(T))$ .

Furthermore, at the intermediate temperature range in which  $\chi(T)$  does not be explained by Eqs. (1) and (3), the theoretical approach for  $Z_s$  is beyond scope of this paper. More detailed discussion with other models will be done in a separate paper.

## VI. CONCLUSION

In conclusion, at  $T_c - T \ll T_c$  (above the condensation temperature), the shielding susceptibilities for BKBO and YBCO crystals well-fit  $\chi(T)/\chi(0) = (1 - exp(-2\Delta(T)/k_B T))$  instead of  $\chi(T)/\chi(0) = (1 - T/T_c)$ . The exponential decrease of the susceptibility indicates that the conduction mechanism is hopping. The calculated surface resistance depends upon the temperature dependence of the number of Cooper pairs at  $T_c - T \ll T_c$ . Furthermore, we contend that the temperature dependence of the diamagnetic shielding susceptibility is that of Cooper pairs;  $\chi(T) \propto n_{sup}(T)$ .

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