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Nonlinear Electrical Conductivity of Superconducting Films below the Transition Temperature

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The Langevin equation for the superconducting order parameter above the transition temperature T_c proposed by Schmid is modified to calculate the nonlinear excess conductivity $\sigma'(T, E)$ slightly below T_c. The electric field dependence of $\sigma'(T, E)$ is described approximately by the same function of $E/E_c(T)$ as Schmid's function above T_c, with a newly defined characteristic field $E_c(T)$ below T_c. The experimental results of the nonlinear electrical conductivity of aluminum films below T_c are in fairly good agreement with the theory.

I. INTRODUCTION

The excess conductivity due to the thermodynamic fluctuations of the superconducting order parameter has been extensively investigated in the temperature region above and below the transition temperature T_c. Smith *et al.*[1] found that the excess current in thin films above T_c shows a nonlinear dependence on the electric field when the velocity v_s of the fluctuation Cooper pairs exceeds $\hbar/m\xi(T)$, where $\xi(T)$ is the temperature-dependent Ginzburg-Landau (GL) coherence length. Since then, the nonlinearity has been studied above T_c: theoretically by Hurault,[2] Schmid,[3] Tsuzuki,[4] and Gor'kov,[5] and experimentally by Thomas and Parks,[6] Klenin and Jensen,[7] and Kajimura and Mikoshiba[8] on thin aluminum films. The experimental results are in qualitative agreement with the theories.

According to the theories,[2–5] the characteristic field $E_c(T)$, at which the nonlinearity becomes appreciable, is proportional to $(T - T_c)^{3/2}$ in the case of thin films above T_c, while the zero-field excess conductivity is proportional to $(T - T_c)^{-1}$. However, the zero-field excess conductivity is observed to be continuous at T_c, and to increase exponentially as the temperature is lowered slightly below T_c. This behavior was successfully explained by Marčelja's theory.[9,10] In this temperature region the nonlinearity is expected to be greatly enhanced, but the theories of Refs. 2–4 cannot be applied to the immediate vicinity of T_c and below T_c. In this paper we present the result of a theoretical and experimental study on the nonlinear excess conductivity of thin films in this temperature region.

In Sec. II, we propose a Langevin equation appropriate for the temperature region slightly below T_c, and calculate the nonlinear excess conductivity. It is shown that the excess conductivity in the zero-electric-field limit reduces to the result given by Masker *et al.*,[9] and the electric field dependence of the excess conductivity is almost the same as that above T_c. The experimental procedure is given in Sec. III. In Sec. IV, we present the experimental results on the excess conductivity as a function of temperature and electric field, and compare them with the present calculation. The agreement between theory and experiment is fairly good. In Sec. V, discussions are given of the validity of the theory in terms of both a phenomenological treatment and a microscopic theory.[11]

II. THEORY

Schmid[3] proposed a Langevin equation for the superconducting order parameter $\Psi(\vec{r}, t)$ above T_c and calculated the nonlinear excess conductivity. Tsuzuki[4] and Gor'kov[5] gave a support to this Langevin equation method by deriving the same result for the excess conductivity using the micro-

scopic theory. We modify the Langevin equation in order to calculate the excess conductivity below T_c retaining the third-order term in Ψ. It takes the form

$$\gamma\left(\hbar\frac{\partial}{\partial t}-2ie\tilde{V}\right)\Psi(\vec{r},t)$$

$$=-\left(\frac{(-i\hbar\vec{\nabla}+2e\vec{A}/c)^2}{2m}-\alpha'\right)\Psi(\vec{r},t)+f(\vec{r},t)\ , \quad (1)$$

where

$$\alpha'=\alpha-\beta'\langle|\Psi(\vec{r},t)|^2\rangle\ . \quad (2)$$

Here $\beta'=2\beta$,[12] and α, β, and γ are the coefficients in the GL equations. The transition temperature T_c is defined as the temperature at which the GL coefficient α vanishes. $\vec{A}$ is the vector potential, $\tilde{V}$ is the electrochemical potential, and $\langle\cdots\rangle$ means the ensemble average. If we neglect the random force $f(\vec{r},t)$, Eq. (1) reduces to the time-dependent GL(TDGL) equation[13] linearized according to Marčelja's idea.[9] The random force $f(\vec{r},t)$ is assumed to be completely uncorrelated in space and time, i.e.,

$$\langle f^*(\vec{r},t)f(\vec{r}',t')\rangle=2\hbar\gamma k_B T\delta(\vec{r}-\vec{r}')\delta(t-t')\ . \quad (3)$$

The prefactor has been chosen in such a way that $\langle|\Psi(\vec{r},t)|^2\rangle$ in the absence of external fields above T_c reduces to the thermodynamic average of $|\Psi(\vec{r},t)|^2$. The validity of the above Langevin equation is discussed in Sec. V and the Appendix.

We consider a system slightly below T_c in a uniform electric field $\vec{E}$. A gauge, $\tilde{V}=0$ and $\vec{A}=-c\vec{E}t$, is chosen. Solving Eq. (1) with respect to the Fourier component of the order parameter, $\Psi_{\vec{q}}=\int d\vec{r}\,\Psi(\vec{r},t)e^{-i\vec{q}\cdot\vec{r}}/V$, V being the volume of the system, and taking the ensemble average of $|\Psi_{\vec{q}}(t)|^2$, we obtain

$$\langle|\Psi_{\vec{q}}(t)|^2\rangle=\frac{2k_BT}{V\hbar\gamma}\int_{-\infty}^{t}dt'$$

$$\times\exp\left[\frac{-2}{\hbar\gamma}\int_{t'}^{t}dt''\left(\frac{(\hbar\vec{q}-2e\vec{E}t'')^2}{2m}-\alpha'\right)\right]. \quad (4)$$

The excess current $\langle\vec{j}(\vec{r},t)\rangle$ is given by

$$\langle\vec{j}(\vec{r},t)\rangle=\sum_{\vec{q}}\frac{-2e}{m}(\hbar\vec{q}-2e\vec{E}t)\langle|\Psi_{\vec{q}}(t)|^2\rangle\ . \quad (5)$$

When we consider the system of a thin film with thickness $d\ll\xi(T)$, we obtain the nonlinear excess conductivity, $\sigma'(T,E)=\langle\vec{j}\rangle/\vec{E}$, as

$$\sigma'(T,E)=\sigma'(T,0)\int_0^\infty du$$

$$\times\exp\left[\frac{\alpha'}{|\alpha_0'|}u-\left(\frac{E}{E_c(T)}\right)^2u^3\right]\ , \quad (6)$$

where

$$\sigma'(T,0)=e^2k_BT\gamma/2\pi\hbar d|\alpha_0'|\ , \quad (7)$$

$$E_c(T)=(3m)^{1/2}|2\alpha_0'|^{3/2}/e\hbar\gamma\ . \quad (8)$$

Here α_0' is the value of α' at $E=0$. Above T_c, the third-order term in Eq. (1) can be ignored and obviously $\alpha'\simeq\alpha$, so that Eq. (6) is reduced to the one obtained by Schmid.[3] In the temperature region slightly below T_c, however, α' depends not only on T but also on E. Then we must determine it by Eq. (2) self-consistently. The quantity $\langle|\Psi(\vec{r},t)|^2\rangle$ in Eq. (2) is obtained from Eq. (4) by summing over $\vec{q}$. Since the naive summation diverges, we introduce the wave-number cutoff taking account of the fact that the GL theory is applicable only to the wave numbers[14] $q<q_c=\xi^{-1}(0)$. Therefore, we introduce a step function $\theta[\hbar q_c-|\hbar\vec{q}-2e\vec{E}t'|]$ into Eq. (4) and sum $\langle|\Psi_{\vec{q}}(t)|^2\rangle$ with respect to $\vec{q}$ in the region[15] $|\hbar\vec{q}-2e\vec{E}t|<\hbar q_c$. Then the self-consistency equation for α', Eq. (2), becomes

$$\alpha'=\alpha-\frac{\beta mk_BT}{\pi\hbar^2d}\int_0^\infty du\,\frac{1}{u}\left[1-\exp\left(\frac{\epsilon_c u}{|\alpha_0'|}\right)\right]$$

$$\times\exp\left[\frac{\alpha' u}{|\alpha_0'|}-\left(\frac{E}{E_c(T)}\right)^2u^3\right]\ , \quad (9)$$

where $\epsilon_c=\hbar^2q_c^2/2m=\alpha(0)$.

In the limit of $E=0$, we have

$$\alpha_0'=\alpha-\frac{\beta mk_BT}{\pi\hbar^2d}\ln\left(1-\frac{\epsilon_c}{\alpha_0'}\right)\ . \quad (10)$$

When $T<T_c$ and $\alpha\gg\xi\equiv\beta mk_BT/\pi\hbar^2d$, we have[16]

$$\alpha_0'\simeq-\epsilon_c e^{-\alpha/\xi}=-\epsilon_c e^{\epsilon T_c/\epsilon_0' T}\ , \quad (11)$$

where $\epsilon=(T-T_c)/T_c$, and[17]

$$\epsilon_0'=[7\zeta(3)e^2/\pi^4\hbar]R_N^{sq}=2.10\times10^{-5}R_N^{sq}\ .$$

$R_N^{sq}=1/d\sigma_N$ and σ_N are the normal-state film resistance per square (Ω/sq) and the normal-state conductivity, respectively. Equation (7) with Eq. (11) gives

$$\sigma'(T,0)=\frac{e^2k_BT\gamma}{2\pi\hbar d\epsilon_c}e^{-\epsilon T_c/\epsilon_0' T}\ . \quad (13)$$

This is essentially the same as is given by Masker *et al.*[9] except for minor differences mentioned in Refs. 16 and 17.

At $E\ll E_c(T)$, we can easily determine α' and we obtain

$$\sigma'(T,E)=\sigma'(T,0)\left[1-\left(6-\frac{2\zeta}{\zeta-\alpha_0'}\right)\left(\frac{E}{E_c(T)}\right)^2\right]\ . \quad (14)$$

The factor $[6-2\zeta/(\zeta-\alpha_0')]$ reduces to 6 far above T_c and 4 below T_c. At $E\gg E_c(T)$, we can neglect the α'-dependent part in Eq. (6), and we have

$$\sigma'(T,E)=S\sigma_N E^{-2/3}\ , \quad (15)$$

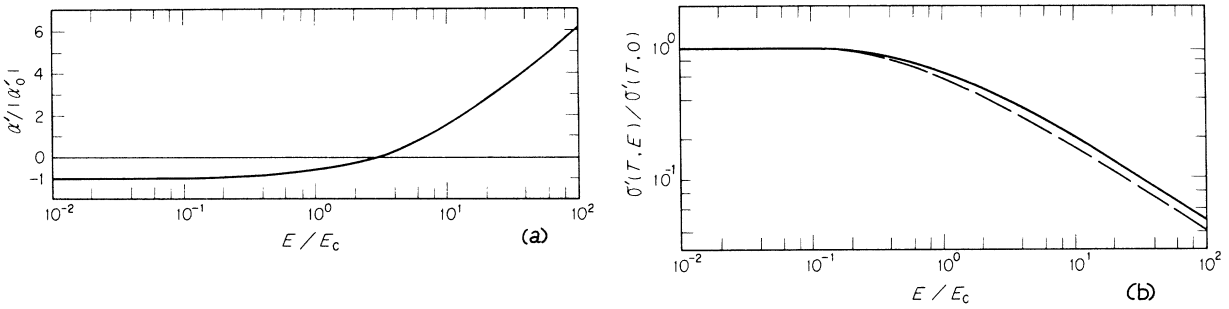

FIG. 1. (a) Electric field dependence of the parameter α' at $T=1.9140$ K calculated by Eq. (9) with parameters of sample A-7. (b) The solid curve shows $\sigma'(T,E)/\sigma'(T,0)$ at $T=1.9140$ K calculated by Eq. (6) with σ' shown in (a). The dashed curve shows $\sigma'(T,E)/\sigma'(T,0)$ calculated by Eq. (17).

with

$$S=\Gamma\left(\frac{4}{3}\right)\frac{e^2k_BT}{\pi\hbar^2d\sigma_N}\left(\frac{3m\hbar\gamma}{e^2}\right)^{1/3} . \quad (16)$$

The $E^{-2/3}$ dependence is the same as that above T_c. At arbitrary electric fields, α' must be determined by numerical calculations.

A result of a numerical calculation of the self-consistency equation for α', Eq. (9), as a function of the electric field is shown in Fig. 1(a) for sample A-7 in Table I at $T=1.9140$ K. The values of $\alpha=1.23\times10^{-19}$ erg, $\beta=2.19\times10^{-37}$ erg cm^3, and $\gamma=0.0398$ are used.[18] Using the values of α', the electric field dependence of the ratio $\sigma'(T,E)/\sigma'(T,0)$ can be calculated by Eq. (6). In Fig. 1(b) the result for $\sigma'(T,E)/\sigma'(T,0)$ is shown by the solid curve against the ratio E/E_c. We can neglect the electric field dependence of $\sigma'(T,E)$ through α', since it is smaller than that through the factor $(E/E_c)^2u^3$ in Eq. (6). In this approximation, $\sigma'(T,E)/\sigma'(T,0)$ can be given by a universal function of E/E_c as

$$\frac{\sigma'(T,E)}{\sigma'(T,0)}=\int_0^\infty du\exp\left[-u-\left(\frac{E}{E_c(T)}\right)^2u^3\right] , \quad (17)$$

which was first derived for the case above T_c with[3] $E_c(T)=(3m)^{1/2}|2\alpha|^{3/2}/e\hbar\gamma$. Equation (17) is shown in Fig. 1(b) by the dashed curve. The difference between the two curves is relatively small, and we make an analysis of the experimental data on $\sigma'(T,E)$ using Eq. (17) in Sec. IV.

III. EXPERIMENTAL PROCEDURE

Films were prepared by evaporation and deposition from an aluminum-wetted coil-type (7-mm diam , 5-cm-long) tungsten filament onto room-temperature slide glass substrates, 20 cm apart from the filament. Thin stainless-steel masks, made by photoetch technique and 0.05–0.15-mm thick, were used to define precisely sample geometries, by which we could avoid film-edge effects.[19] The size of the tapered fringe part of the prepared films was estimated to be 1 μm. The dirty films were deposited at a rate of 5 Å/sec and at oxygen pressures of 10^{-4} Torr, and their dimensions were 1.0 mm wide and 10.0 mm long. The clean films were prepared at a deposition rate of 50 Å/sec in vacua of $(1-15)\times10^{-6}$ Torr, and were rectangular zig-zag patterns having a large length-to-width ratio, 0.2 mm$\times$20 cm. The possibility of nonuniformity in the samples was reduced by confining the patterns to an area of the film less than 0.8 cm^2. Maximum thickness variation in one sample was estimated to be $\frac{1}{200}$. After deposition of samples, films were coated with paraffin to avoid the oxidation with the laboratory air. Film thickness was measured with a multiple-beam interference microscope within an accuracy of $\pm$20 Å.

The resistance of Al samples was measured in the temperature range 4.3–1.4 K by measuring the dc or ac voltage across the films prepared for the conventional four-terminal method. A five-decade digital voltmeter with an accuracy of 0.1 μV was used to measure the voltage across the films and across a standard resistance to monitor the current in series with a sample. The measured dc currents, which ranged from 0.01–300 μA, were provided by fifteen 1.5-V dry cells with high resistance in series with a film and were constant to

TABLE I. Observed values of sample parameters.

Sample	R_N^{sq} (Ω/sq)	d (Å)	$\xi(0)$ (Å)	$10^3\epsilon_0'$
A-3	11.531	120	1000	0.143
A-4	37.727	160	650	0.553
A-7	128.45	100	500	1.13
A-8	129.12	100	480	1.15
A-9	166.90	280	240	1.00
A-10	238.24	100	450	2.41
A-11	374.61	100	280	7.75
A-12	869.03	200	130	7.85
A-13	1999.2	140	80	12.7

within one part in 10^5 over 1 h. The electric field dependence of the excess conductivity was measured by the dc digital voltmeter for two samples. For other samples, a phase-sensitive detector was used to measure the resistance by supplying a small constant ac current with a range of 0.01–0.1 μA for small voltages across the samples. The magnitude of the electric field was two orders of magnitude smaller than the characteristic field $E_c(T)$ in Eq. (8), and the measured excess conductivity was regarded as the zero-field value.

The temperature of samples which were immersed in liquid helium was determined by measuring the vapor pressure within an accuracy of ± 0.1 Torr in the range 770–80 Torr, and of ± 5 mTorr in the range 80–2 Torr. The relative accuracy of the temperature is about 0.05 mdeg, and the absolute one is 1 mdeg. Maximum temperature difference between samples and the liquid-helium bath caused by sample heating due to the Joule loss and the Kapitza resistance was estimated to be 1.2 mdeg in the electric field of about 200 mV/cm. When the electric field is lowered, the temperature difference due to the Joule loss rapidly decreases.

The samples were mounted with the sample surface parallel to the Dewar axis. All the measurements without external magnetic fields were carried out in annealed Mu-metal shields within which the earth's magnetic field was reduced to 8×10^{-4} Oe. But the field component perpendicular to the sample surface can be reduced to less than 10^{-4} Oe by rotating the samples around the Dewar axis.

It is very important to determine precisely the normal-state resistance R_N of films in order to obtain the accurate electric field dependence of $\sigma'(T,E)$. The normal-state resistance R_N was determined from the measurement of the magnetic field dependence of the sample resistance with the help of the formula given by Abrahams *et al.*[20] and Maki[21] for the excess conductivity in the perpendicular magnetic field H, i.e.,

$$\sigma'(T,H)=\sigma'(T,0)\,2\left(\frac{\epsilon}{h}\right)^2 \times\left[\psi\left(\frac{1}{2}+\frac{\epsilon}{2h}\right)-\psi\left(1+\frac{\epsilon}{2h}\right)+\frac{h}{\epsilon}\right], \quad (18)$$

where $h=2\pi\xi^2(0)H/\phi_0$, ϕ_0 is the flux quantum, $\sigma'(T,0)=e^2/(16\hbar d\epsilon)$, and ψ is the digamma function. The formula reduces to

$$\frac{1}{R(H)}-\frac{1}{R_N}=\frac{e^2}{8\hbar d\sigma_N}\,\frac{1-(2\epsilon/h)\ln 2}{hR_N}, \quad (19)$$

when $|\epsilon/h|\ll 1$. Then by plotting $1/R(H)$ vs $1/H$ at $\epsilon\simeq 0$, the data were extrapolated to $1/H=0$, yielding a value of R_N. In this case, the Maki term[22] is negligible because the depairing effect in the high perpendicular magnetic field is very large.

The value of the GL coherence length at 0 K, $\xi(0)$, was deduced from the measurement of the critical field as a function of temperature in the perpendicular magnetic field, $H_{c1}(T)$, which is $[\phi_0/2\pi\xi^2(0)]|\epsilon|$ in the vicinity of T_c.[23] In the dirty limit, we can use the Gor'kov theory[24] to express the GL coherence length in terms of the electronic mean free path l and the BCS coherence length[25] ξ_0 as $\xi^2(0)=0.72\xi_0 l$.

By this means, parameters of Al films prepared in the above-mentioned manner were determined and are tabulated in Table I.

IV. EXPERIMENTAL RESULTS

In this section we present the experimental results on the electric field dependence of the excess conductivity $\sigma'(T,E)$ at various constant temperatures and the temperature dependence of the excess conductivity in the zero-electric-field limit below T_c. The results are compared with the calculation in Sec. II.

The quantity $\log_{10}[\sigma'(T,E)/\sigma_N]$ for an aluminum film A-7 with $d=100$ Å and $R_N^{sq}=128.45$ Ω/sq at various constant temperatures T is plotted against $\log_{10}E$ in Fig. 2. It clearly shows the $E^{-2/3}$ law, Eq. (15), at large fields with the observed value $S_{obs}=18$ (mV/cm)$^{2/3}$. On the other hand, we obtain $S_{calc}=7.9$ (mV/cm)$^{2/3}$ using γ calculated from the microscopic expression, the parameters mentioned in Sec. III and the value $T\simeq T_c=1.9191$ K.[26] The agreement is fairly good. In the framework of the present theory the small discrepancy can only be attributed to the smallness of the theoretical value of $\gamma=0.0398$ compared with $\gamma=0.48$ reduced from S_{obs}. The $E^{-2/3}$ dependence is the same as that observed by Thomas and Parks,[6] and Klenin and Jensen[7] at temperatures above T_c.

The solid curves in Fig. 2 are the theoretical ones calculated by Eq. (17). In this figure all the curves for various temperatures have the same form, only shifted from each other along the temperature-independent line of Eq. (15). The fit is good over five orders of magnitude of the electric field. The deviation of experimental points from the theory for $T=1.9150$ K and $T=1.9219$ K is not due to the sample heating. The deviation occurs at rather low electric fields, where the sample-bath temperature difference is estimated to be about 0.05 mdeg, as has been discussed in Sec. III. This type of behavior is seen in other pure samples[27] at temperatures near T_c. The same electric field dependence as in Fig. 2 has been observed[6,8,27] above T_c.

The zero-field excess conductivity $\sigma'(T,0)$ is determined by the above-mentioned fit, and the

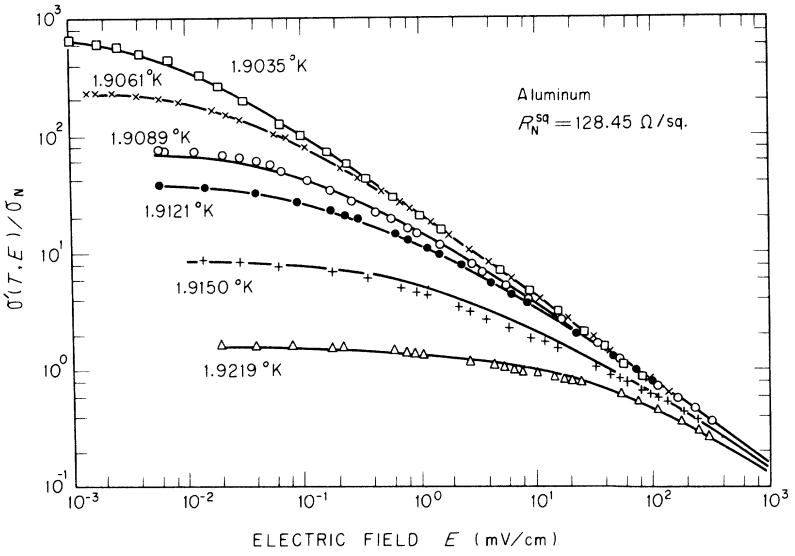

FIG. 2. Variation of the quantity $\sigma'(T, E)/\sigma_N$ of a superconducting aluminum film A-7 ($d = 100$ Å, $R_N^{sq} = 128.45$ Ω/sq) with the electric field at various constant temperatures ($T_c = 1.9191$ K). Solid curves show the theoretical values of Eq. (17). All the curves for various temperatures have the same form, and are fitted to the data by shifting each other.

quantity $\sigma_N/\sigma'(T, 0)$ of the same specimen is plotted against the temperature T in Fig. 3. The same kind of plot for sample A-4 is shown in Fig. 4. In this case $\sigma_N/\sigma'(T)$ was measured with a small constant ac-current (0.01 μA) method, mentioned in Sec. III. Below T_c, $\sigma'(T, 0)$ is given by Eq. (13). The exponential behavior of $\sigma'(T, 0)$ is clearly observed in both plots. The temperature dependence of $\sigma'(T, 0)$ is similar to that observed by Masker *et al.*[9] The observed values of ϵ_0' are plotted against R_N^{sq} in Fig. 5 together with the data obtained by Masker *et al.* The present values of ϵ_0' are about a half of the theoretical values and are about two times the ones obtained by Masker *et al.* The temperature region and the range of the resistive transition of our experiment are as wide as in Ref. 9(c). We have rechecked the procedure to measure the temperature and the resistance but reaffirmed the accuracy described in Sec. III. As has been discussed by Masker *et al.*, the magnetic field has a large effect on cleaner samples and can lead to the apparently larger values of ϵ_0'. In order to check whether shielding from the earth's field is sufficient, the samples were rotated around the Dewar axis. This procedure could reduce the earth's field perpendicular to the sample surface to less than 10^{-4} Oe. However, we have detected no effect. At present we have no explanation of the discrepancy in the observed values of ϵ_0'. Masker *et al.* argued in a note added in proof in Ref. 9(c) that the discrepancy between the values of ϵ_0' in their theory and experiment is reduced to a factor of 1.1, if they take into account the deviation of ρl_{eff} from the free-electron value due to the size effect, where ρ and l_{eff} are the resistivity and the electronic mean free path, respectively. However, we think that the deviation of ρl_{eff} may be included in the measured values of R_N^{sq}, and therefore the discrepancy between the values of ϵ_0' cannot be reduced.

V. DISCUSSIONS AND SUMMARY

The unlinearized TDGL equation augmented by the random force $f(\vec{r}, t)$ with the property given by Eq. (3) is more general and more appropriate than the linearized Langevin equation used in this paper. The reason is that in the absence of time-dependent external fields the equivalent Fokker-Planck equation has a stationary solution,[28, 29] $\exp\{-F[\Psi(\vec{r})]/k_B T\}$, where $F[\Psi(\vec{r})]$ is the GL free energy. McCumber and Halperin solved this

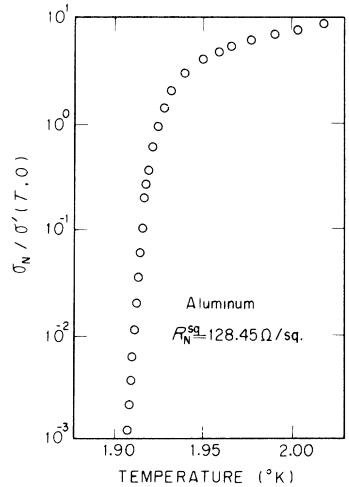

FIG. 3. Temperature dependence of the quantity $\sigma_N/\sigma'(T, 0)$ of sample A-7 determined from the fit in Fig. 2. For the sake of clarity, only a small fraction of the data points are shown.

Fokker-Planck equation for thin wires in the temperature region where the nonlinearity in $\Psi(\vec{r}, t)$ is essential.[28, 30] On the other hand, our Langevin equation (1) with Eq. (2) can be considered as the one obtained by linearizing the unlinearized Langevin equation. The validity of the linearization is examined in the case of thin films in the absence of external fields in the Appendix. It is shown that the linearization is valid when

$$\epsilon > \eta_c = \epsilon_0'(1 + \ln\epsilon_0') \,, \tag{20}$$

where ϵ_0' is defined by Eq. (12). In the case of sample A-7, $(\epsilon_0')_{\text{calc}} \simeq 2.7 \times 10^{-3}$ and $\eta_c \simeq -1.3 \times 10^{-2}$, and the above region covers the region where $\sigma'(T, 0)$ is observed to increase exponentially.

Precisely the same result as Eqs. (6) and (9) can be obtained by a microscopic theory.[11] This theory is based on the expression for the excess current and the Dyson equation for the fluctuation propagator, in imaginary time and in an external field, as is given by Tsuzuki.[4] The Dyson equation is slightly modified so that it pertains not only to the interaction with external fields, but also to the interaction between fluctuation propagators self-consistently in the lowest order.[31] From the direct analytical continuation from the imaginary to real time, precisely the same equations as Eqs. (6) and (9) are derived. When we consider the validity of this self-consistent treatment for the fluctuation propagator, the same criterion as Eq. (20) is obtained. The details are shown in a separate paper.[11]

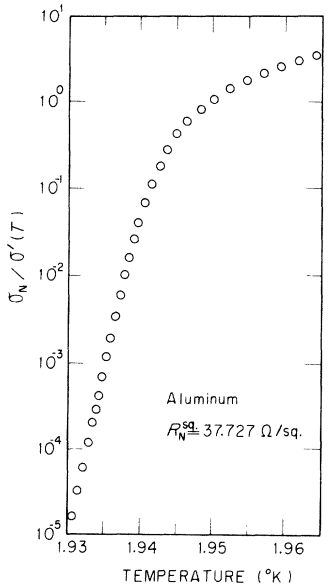

FIG. 4. Temperature dependence of the quantity $\sigma_N/\sigma'(T, 0)$ of sample A-2 ($d = 160$ Å, $R_N^{sq} = 37.727$ Ω/sq). The quantity $\sigma_N/\sigma'(T)$ was measured with a small constant ac-current method mentioned in Sec. III.

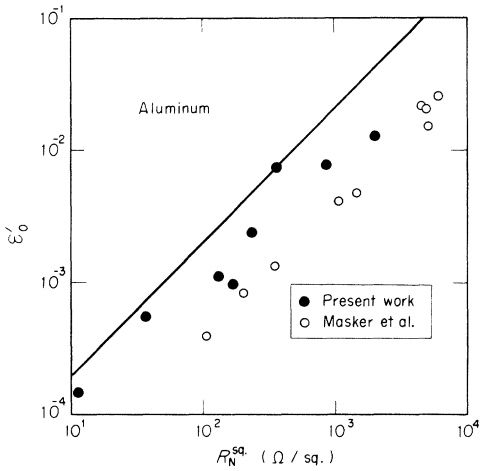

FIG. 5. A log-log plot of ϵ_0 vs R_N^{sq}. The solid line shows the theoretical value in Eq. (12).

In both the present calculation and the microscopic theory we neglect the contribution from the so-called Maki term[22] or the anomalous term[32] in the basic equations. Since the Maki term is considered to give the change in the normal current due to the fluctuation Cooper pairs, its contribution seems to be negligible at temperatures below T_c where the current carried by Cooper pairs is dominant. Actually, the zero-field contribution from the Maki term can be shown to be negligible in the self-consistent microscopic theory. Recently, however, Maki[33] argued that the Maki term becomes twice as large as the value of Eq. (15) at $E \gg E_c$ and temperature above T_c. If this is true, it may explain the discrepancy between S_{obs} and S_{calc} in our case below T_c. Thus, the Maki-term problem must be studied further.

In summary, we have modified the Langevin equation and calculated the nonlinear excess electrical conductivity $\sigma'(T, E)$ slightly below T_c. The zero-field conductivity $\sigma'(T, 0)$ is essentially reduced to the result given by Masker *et al.* The electric field dependence of the normalized value $\sigma'(T, E)/\sigma'(T, 0)$ is described approximately by the same function of $E/E_c(T)$ as that of Schmid above T_c, where $E_c(T)$ is the newly defined characteristic field. The experimental result on aluminum films is in fairly good agreement with the calculation on three points: (i) the $E^{-2/3}$ dependence of $\sigma'(T, E)$ at high fields, (ii) the electric field dependence of $\sigma'(T, E)/\sigma'(T, 0)$ at intermediate fields, and (iii) the exponential temperature dependence of $\sigma'(T, 0)$.

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APPENDIX

The unlinearized TDGL equation augmented by the random force $f(\vec{r},t)$ with the property given by Eq. (3) can be rewritten in the form of Eq. (1) in which, however, $f(\vec{r},t)$ should be replaced by a new random force $K(\vec{r},t)$ defined by

$$K(\vec{r},t)=f(\vec{r},t)+\beta(|\Psi|^2-2\langle|\Psi|^2\rangle)\Psi(\vec{r},t)\ . \qquad \text{(A1)}$$

We shall see below that at temperatures slightly below T_c the second Ψ-dependent part of $K(\vec{r},t)$ makes negligible correction to the contribution from the first term in the absence of external fields and that the nonlinear Langevin equation is plausibly reduced to the linearized form in Eq. (1) in a certain temperature region.

If we assume that $f(\vec{r},t)$ is a Gaussian random variable, we can obtain the correction to any correlation function of Ψ and Ψ^* due to the Ψ-dependent part of $K(\vec{r},t)$ by iteration. We obtain the average $\langle|\Psi(\vec{r},t)|^2\rangle$ for a thin film in the absence of external fields as

$$\langle|\Psi(\vec{r},t)|^2\rangle=\frac{1}{V}\sum_{\vec{q}}\frac{k_BT}{\epsilon_q-\alpha_0'}+\frac{1}{V^3}\sum_{\vec{q}_1,\ldots,\vec{q}_4}\frac{2\beta^2(k_BT)^3\delta_{\vec{q}_1+\vec{q}_2,\vec{q}_3+\vec{q}_4}}{(\epsilon_{q_1}-\alpha_0')(\epsilon_{q_2}-\alpha_0')}\frac{1}{(\epsilon_{q_3}-\alpha_0')(\epsilon_{q_4}-\alpha_0')(\epsilon_{q_1}+\epsilon_{q_2}+\epsilon_{q_3}+\epsilon_{q_4}-4\alpha_0')}+\cdots$$

$$=\frac{\zeta}{2\beta}\ln\left|\frac{\epsilon_c-\alpha_0'}{\alpha_0'}\right|+\frac{\nu\zeta^3}{2\beta|\alpha_0'|^2}+\cdots\ , \qquad \text{(A2)}$$

where $\epsilon_q=\hbar^2q^2/2m$, ζ is defined below Eq. (10), and

$$\nu=\frac{1}{2\pi^3}\int\frac{d^2r_1d^2r_2d^2r_3d^2r_4\,\delta(\vec{r}_1+\vec{r}_2-\vec{r}_3-\vec{r}_4)}{(r_1^2+1)(r_2^2+1)(r_3^2+1)(r_4^2+1)(r_1^2+r_2^2+r_3^2+r_4^2+4)}\simeq 0.17\ .$$

Equation (2) with Eq. (A2) gives the self-consistency condition for α_0',

$$\alpha=\alpha_0'+\zeta\ln\left|\frac{\epsilon_c-\alpha_0'}{\alpha_0'}\right|+\frac{\nu\zeta^3}{|\alpha_0'|^2}+\cdots\ . \qquad \text{(A3)}$$

The third- and higher-order terms in the right-hand side of this equation become important when $|\alpha_0'|$ becomes nearly equal to ζ. In the temperature region where the condition

$$\alpha<-\zeta+\zeta\ln[(\epsilon_c+\zeta)/\zeta] \qquad \text{(A4)}$$

is satisfied, we can neglect the third- and higher-order terms, i.e., the contribution from the Ψ-dependent part of $K(\vec{r},t)$. The condition (A4) reduces to Eq. (20) when we use the microscopic expressions for α and β.

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for the t' integration is released to $-\infty$. The error caused by these procedures is estimated to be bounded by the order $eE\gamma/q_c\epsilon_c$ and is negligible. On the other hand, the current expression is convergent without the restriction.

[16]Equation (11) can be obtained from Eq. (15) in Ref. 9(c) apart from a factor of 2 caused by the difference between β and β', if we assume $\epsilon_c \ll k_B T$, as is the case for ordinarily dirty materials. In this case, the Bose character assumed by Masket *et al.* makes no effects. Although when $\epsilon_c \gg k_B T$ (which may occur in the case of extremely dirty materials), the assumed Bose character leads to Eq. (16) in Ref. 9(c), we will not make this assumption; therefore, Eq. (11) is correct in both cases.

[17]The magnitude of the value of ϵ_0' in Ref. 9(c) is half that of the present value. The factor of 2 in our expression is due to the use of β' instead of β.

[18]R_N^{sq} and $\xi(0)$ are given in Table I, and $T_c = 1.9191$ K. The quantity $\alpha(0)$ is determined by $\alpha(0) = \hbar^2/2m\xi^2(0)$, and γ by $\gamma = \pi\alpha(0)/8k_B T_c$. To determine β we use $\beta = 1.02\hbar^2/nl^2m$ where $n = 5.56 \times 10^{22}$ cm^{-3} and $l = 32$ Å (Ref. 25). α_0' can be obtained from Eq. (7) with the above-mentioned values of T_c and γ, and α from Eq. (10) with the values of α_0' and β. This value of α is in plausible agreement with the value obtained by $\alpha = -\alpha(0)\epsilon = 7.3 \times 10^{-2}$ erg.

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Antiferromagnetic Solution of the Hubbard Model*

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The Hubbard Hamiltonian for the system of one-electron atoms is solved in the presence of sublattice magnetization. In the limit of the fully antiferromagnetic state, the results reproduce those of Slater's split-band model by splitting a nonmagnetic band into spin-polarized bands. As magnetization decreases, antipolarized split bands appear and increase their strengths while the band gap remains constant. In the limit of no sublattice magnetization, the strengths of the two types of bands become equal, yielding the Hubbard nonmagnetic insulating state.

I. INTRODUCTION

Many of transition-metal oxides are good insulators, even though their d band is only partly filled. At low temperatures, they often exhibit some antiferromagnetic spin ordering but remain insulating well above the Néel temperatures where the spin ordering has completely disappeared.[1] An insulating antiferromagnetic state may be well described by Slater's split-band model,[2] but the band gap involved is proportional to the sublattice magnetization and hence vanishes as the antiferromagnetic spin ordering disappears. Therefore, this model is incapable of describing the insulating state of transition-metal oxides properly.

Hubbard[3,4] suggested that such an insulating state is a consequence of strong correlation effects in d bands. He further argued that, since the intra-