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Strong reduction of quasiparticle scattering rate with gap formation in CeNiSn

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We measured the temperature dependence of the surface impedance $Z_s = R_s + iX_s$ at 9.6 and 14 GHz in a single crystal of the Kondo lattice compound CeNiSn, from which microwave complex conductivity $\sigma(T) = \sigma_1(T) - i\sigma_2(T)$ was determined. Below about 10 K the imaginary part σ_2 increases rapidly and the real part σ_1 is much lower than the dc conductivity. These results indicate a rapid decrease in the scattering rate of the quasiparticles, whose excitation spectrum is drastically changed with the coherence gap formation at low temperatures. [S0163-1829(97)04936-9]

I. INTRODUCTION

Among Kondo-lattice compounds, which have local magnetic moments due to $4f$ or $5f$ electrons at periodic sites, so-called ''Kondo semiconductors'' such as CeNiSn and CeRhSb have attracted much attention. They show a characteristic crossover from metallic heavy-fermion to semiconductinglike or semimetalliclike behavior with the formation of a small energy gap in the electronic excitation spectrum below about 10 K.[1–4] The temperature dependence of the NMR relaxation rate has been explained by a pseudogap with V-shaped structure, which is quite different from a usual activation type.[3] This pseudogap is considered an anisotropic gap, which vanishes in a direction in the momentum space. Tunneling spectrum measurements have also revealed clearly the occurrence of a gap near the Fermi surface of the order of 10 meV in CeNiSn.[4] The magnetoresistance measurements have suggested that the gap is reduced by application of a magnetic field along the a axis of the crystal.[5–7] In addition, the inelastic neutron scattering studies show the existence of anisotropic magnetic excitations.[8,9]

The transport properties of CeNiSn are somewhat confusing. Clean CeNiSn single crystals recently obtained have exhibited completely metallic temperature dependence of resistivity along the orthorhombic a axis down to 1.4 K.[5,6] Only poor samples with some impurity phases become semiconductinglike. For the latter, the magnitude of the Hall coefficient was reported to be enhanced at low temperatures, which seemed consistent with a gap formation. However, such an effect occurs even in metallic samples.[6,10] The question is why the resistivity shows metallic behavior even if a gap is formed at low temperatures. In order to elucidate such unusual transport properties, it is very important to extract the scattering time of carriers, since in a simple model the resistivity is a function of the effective mass m^*, the carrier number n, and the (renormalized) scattering time τ; $\rho(T) = m^*/ne^2\tau$.

Measurements of microwave complex conductivity $\sigma = \sigma_1 - i\sigma_2$ are powerful to investigate the scattering time τ.[11–15] The real and imaginary parts of the conductivity vary differently as functions of τ. If we can determine σ_1 and σ_2 simultaneously, a quantitative analysis of τ becomes possible. In contrast to the optical reflectivity measurements, which need Kramers-Kronig transformations to extract σ_1 and σ_2, the microwave cavity perturbation technique can directly determine these quantities at a fixed frequency. We have measured the surface impedance and determined the temperature dependences of $\sigma_1(T)$ and $\sigma_2(T)$ at 9.6 and 14 GHz in a CeNiSn single crystal. Using the Drude model, we found that the scattering time of the carriers is drastically enhanced below about 10 K. This result indicates a strong reduction of the scattering rate with the formation of the gap in the quasiparticle spectrum.

II. EXPERIMENT

The CeNiSn single crystal used in the present study was grown from stoichiometric starting materials by a Czochralski method.[5] Similar crystals were characterized previously by magnetic susceptibility, transport, and thermodynamic measurements.[5,6] The crystal used in this study is of relatively high purity. The impurity phases (Ce_2O_3 and $CeNi_2Sn_2$) were found to be less than 0.2%. It shows metallic behavior along the a axis down to 1.4 K, and is semiconductive in other directions [see Fig. 1(a)]. For the microwave surface impedance measurements, the crystal was cut into a parallelepiped with the dimensions of 1.16, 1.03, and 1.30 mm along the a, b, and c axes, respectively.

The temperature dependence of the surface impedance was measured by using a superconducting cavity reson-

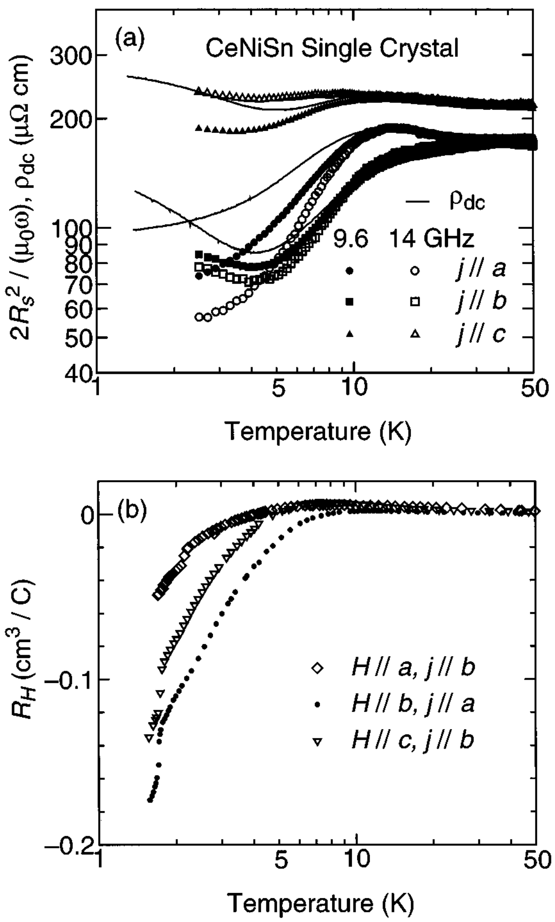

FIG. 1. (a) Temperature dependence of $2R_s^2/(\mu_0\omega)$ at 9.6 and 14 GHz along the a, b, and c directions in the CeNiSn single crystal. The solid curves represent the temperature dependence of anisotropic resistivity $\rho_{dc}(T)$. (b) Temperature dependence of Hall coefficient in a crystal which shows similar $\rho_{dc}(T)$ (after Ref. 7).

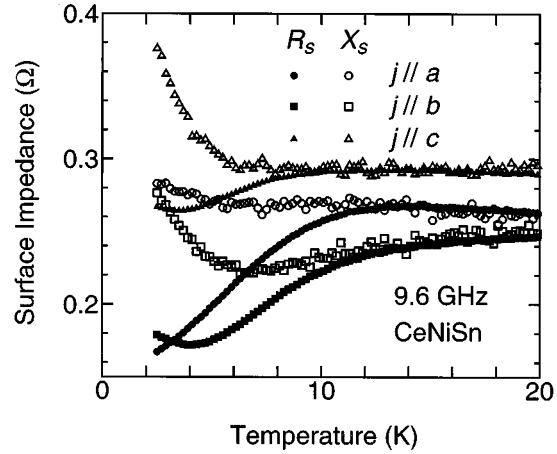

FIG. 2. Temperature dependence of surface resistance R_s and surface reactance X_s at 9.6 GHz. In all the directions, X_s (open symbols) deviates from R_s (closed symbols) below about 10 K.

ator.[13,15,16] The cavity is maintained at liquid He temperature, while the sample on a sapphire rod can be heated up to 100 K. The sample is placed in the center of the cylindrical cavity, where the magnetic field component H_{ac} of microwave is maximum along the cavity axis for both TE_{011} and TE_{013} resonance modes. Therefore, we can measure the resonant frequency f and Q at two frequencies in one experimental run. The cavity showed sharp resonances ($Q \sim 10^6$) at 9.6 GHz for the TE_{011} mode and at 14.1 GHz for the TE_{013} mode. The real and imaginary parts of the surface impedance $Z_s = R_s + iX_s$ are determined by the change in Q^{-1} and f, respectively.[13,15,16] In the normal skin effect regime [where the skin depth $\delta_{cl} = (2/\mu_0\omega\sigma)^{1/2}$ is much longer than the mean free path l],[17] the surface impedance is related to the complex conductivity $\sigma = \sigma_1 - i\sigma_2$ by

$$Z_s = R_s + iX_s = \left(\frac{i\mu_0\omega}{\sigma_1 - i\sigma_2}\right)^{1/2}, \tag{1}$$

where μ_0 is the permeability of the vacuum and $\omega = 2\pi f$. Thus, at high temperatures where the Hagen-Rubens relation holds ($\sigma_1 \gg \sigma_2$ and $\sigma_1 \simeq \sigma_{dc}$), we can derive

$$R_s = X_s = (\mu_0\omega/2\sigma_{dc})^{1/2}. \tag{2}$$

Using this equation, $2R_s^2/\mu_0\omega$ should be equal to the dc resistivity ρ_{dc} at high temperatures.

For the measurements of the anisotropy of Z_s, the crystal was oriented with the a, b, and c axes parallel to H_{ac}.[18–20] For example, in the configuration of $H_{ac} \| a$ the ac eddy current flows in the b-c plane. In this case, the measured surface impedance is expressed as $(L^b Z_s^b + L^c Z_s^c)/(L^b + L^c)$, where L^i is the appropriate dimension of the crystal and the superscript i ($i = a$, b, or c) denotes the direction of the current. If we measure the three configurations, we can determine $Z_s^i(T)$ ($i = a$, b, or c) and hence $\sigma^i(T)$ through Eq. (1).

III. RESULTS AND DISCUSSION

Figure 1(a) shows the temperature dependence of $2R_s^2/\mu_0\omega$ with the current j flowing along each direction in the single crystal of CeNiSn. Also shown are the data of anisotropic dc resistivity $\rho_{dc}(T)$ measured by using crystals from the same batch (No. 3 of Ref. 6), and in Fig. 1(b) are the Hall coefficient data for crystal No. 4 of Ref. 6 which shows similar $\rho_{dc}(T)$. At temperatures above 10 K, the dc data and the microwave data coincide with each other. This means that the Hagen-Rubens relation [Eq. (2)] holds in this temperature range. At low temperatures, however, $2R_s^2/\mu_0\omega$ deviates from the dc resistivity (solid curves). Except for $j \| c$ at 14 GHz, higher frequency data show smaller values. In Fig. 2, the surface resistance R_s below 20 K is compared with the surface reactance X_s in a linear scale. It clearly shows that below about 10 K the relation $R_s = X_s$ [Eq. (2)] breaks down, and the difference $X_s - R_s$ increases as the temperature is lowered. These results indicate that at low temperatures we cannot ignore the imaginary part σ_2 of the complex conductivity in the denominator of Eq. (1) in this microwave frequency range. These results are different from those of millimeter-wave measurements reported for $CePd_3$ polycrystals,[11,12] from which the authors claimed that the contribution of σ_2 was small.

Using Eq. (1), we can directly determine the temperature dependence of the real and imaginary parts of the conductivity $\sigma = \sigma_1 - i\sigma_2$ of our CeNiSn single crystal. Figure 3

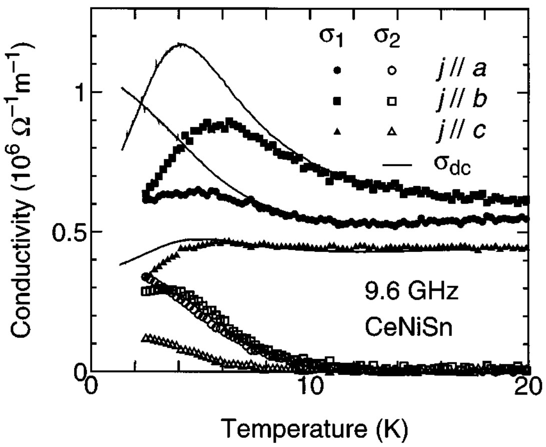

FIG. 3. Real and imaginary parts of complex conductivity $\sigma=\sigma_1-i\sigma_2$ at 9.6 GHz as functions of temperature in the CeNiSn single crystal. The solid curves are the data of dc conductivity.

shows the low-temperature data for $\sigma_1(T)$ and $\sigma_2(T)$ in comparison with the dc conductivity. In this figure we can see two interesting points. One is the anomalously large enhancement of σ_2 (open symbols), which increases to the same order as σ_1 (closed symbols). The second point is the considerable deviation of σ_1 from the dc conductivity at low temperatures. These features can be interpreted by a simple analysis. Let us consider the single-band Drude model for low-frequency conductivity. In this model, the complex conductivity is expressed as

$$\sigma_1-i\sigma_2=\frac{ne^2\tau/m^*}{1+(\omega\tau)^2}-i\frac{\omega\tau\cdot ne^2\tau/m^*}{1+(\omega\tau)^2}, \tag{3}$$

where n and m^* are the number and mass of the carriers, and τ is the scattering time. In this heavy-fermion material, the simple Drude model cannot be a complete expression for high-frequency (optical) response. In experimental analysis, however, the Drude model can be a good description of low-frequency (microwave) conductivity particularly for metallic samples. In other words, we can replace conductivity problem with the scattering rate using the Drude model. If the scattering time is drastically enhanced at low temperatures, the observed enhancement of σ_2 and the suppression of σ_1 are naturally consistent with Eq. (3). Since we did not use exactly the same crystal in the microwave and dc resistivity measurements, the quantitative comparison might be dangerous at low temperatures. Nevertheless, we can estimate the scattering time at the fixed frequencies without using the dc data. From Eq. (3) one can easily derive

$$\tau=\sigma_2/(\sigma_1\omega). \tag{4}$$

The right-hand side of Eq. (4) is the measured quantity in our microwave experiment. Thus, the observed enhancement of σ_2 indicates a rapid increase in scattering time τ.

The temperature dependence of $\sigma_2/(\sigma_1\omega)$ at 9.6 and 14 GHz is plotted in Fig. 4. For all the directions, $\sigma_2/(\sigma_1\omega)$ is drastically enhanced below about 10 K, indicating a strong increase in the scattering time τ in a single-band Drude model. The magnitude of $\omega\tau$ becomes of the order of unity at low temperatures, which means that the contribution of the denominator of Eq. (3) is not negligible. Particularly in the a and b directions, the absolute values of $\sigma_2/(\sigma_1\omega)$ at the two frequencies coincide with each other within the experimental errors. This means that the simple Drude model can explain the microwave electrodynamics along the a and b axes in this material.

Turning to the transport properties, the resistivity and the Hall coefficient have completely different temperature dependence (see Fig. 1). This simply can now be attributed to the fact that τ/m^* is not constant in temperature. Therefore, the observed strong temperature dependence of τ is consistent with transport properties. Then the next question is why the scattering time τ grows up so rapidly below about 10 K. When we look at the quasiparticle excitation spectrum, the density of states near the Fermi level decreases rapidly when the gap is formed.[4] Such a decrease in the density of states is also confirmed in pure metallic samples by the recent specific heat[6] and NMR measurements.[21] If the system has a strong electron-electron correlation, the quasiparticle scattering rate $1/\tau$ should be strongly reduced with the gap formation, because the amount of scattering decreases. In other words, the observed enhancement of τ (reduction of the scattering rate $1/\tau$) of the quasiparticles strongly suggests that the scattering of the quasiparticles has some kind of electronic origin, and is reduced when the density of states near the Fermi level decreases due to the gap formation. This is in clear contrast to the case of usual band-gap semiconductors.

An enhancement of τ has been also reported in the heavy-fermion superconductor UPt_3 and in the intermediated valence compound $CePd_3$ by using a Drude analysis.[11,12] In these compounds, however, the temperature dependence of $1/\tau$ at low temperatures is the same as that of the resistivity, indicating that n/m^* is independent of temperature. In our case, the scattering rate $1/\tau$ in CeNiSn shows much more drastic reduction below 10 K while the resistivity has weak temperature dependence. This shows n/m^* also decreases rapidly at low temperatures in contrast to the case of UPt_3 and $CePd_3$. The above difference can support the gap formation in the CeNiSn quasiparticle spectrum, as discussed in the previous paragraph.

It is very interesting to point out that a reduction of the scattering rate $1/\tau$ has been reported[13,14] also in high-T_c superconductors when the superconducting gap opens in this case. In the superconducting state, the microwave conductivity $\sigma_1(T)$ is enhanced below T_c showing a broad maximum and then decreases as T goes to zero. This enhancement in $\sigma_1(T)$ is much larger than that predicted by the ordinary BCS theory, and has been attributed to the effect of decreasing scattering rate of the quasiparticles in the superconducting state. At low temperatures $\sigma_1(T)$ does not show activated behavior, which is inconsistent with an isotropic gap as it exists in the ordinary s-wave superconductors.[14,22] In CeNiSn, $\sigma_1(T)$ shows a similar broad maximum[23] and nonactivated behavior at low temperatures (see Fig. 3). This result implies that this material has an anisotropic gap and low energy quasiparticle excitations just like d-wave superconductors. Another similarity between CeNiSn and high-T_c superconductors can be seen in the thermal conductivity data.[24,25] The temperature dependence of the thermal conductivity in CeNiSn shows a maximum at around 5 K,[24] similar to the maximum observed for T less than T_c in the thermal conductivity of high-T_c cuprates.[25] Although the phonon and the electron contributions to the thermal conductivity cannot be separated, this maximum could be attributed simply to an increase in phonon conductivity. However, it may be due also to the interplay of decreasing scattering rate and decreasing number of carriers.

Now the transport properties[5,6,10] can be tentatively explained as follows. As mentioned above, the specific heat results[5,6] show a strong decrease of C/T below about 6 K, suggesting a strong decrease in carrier number with the gap formation. If the decrease in the scattering rate is larger than the change in carrier number, the resistivity may exhibit metallic behavior as observed in clean samples. In a dirty sample, however, the scattering rate remains high by a contribution from impurity-scattering $1/\tau_{\mathrm{imp}}$ at low temperatures, and the resistivity shows semiconductive upturn mainly due to the decrease in carrier number. Recently, such impurity-dependent transport properties were theoretically investigated by Ikeda and Miyake,[26] considering the momentum dependence of the hybridization between f and conduction electrons. They claimed that an anisotropic hybridization gap is formed by the highly renormalized quasiparticles near the Fermi level, which is consistent with the experimental investigations.

There still remain, however, some open questions on the transport properties of CeNiSn. One needs to explain the complex behavior of the magnetoresistance[5–7] (closing of the gap by a magnetic field along the a axis) and whether these effects are related to the occurrence of dynamical antiferromagnetic correlations at low temperatures.[8,9] The Hall effect should be measured below 1.5 K and in high field, in order to check whether there could be any simple relation between the carrier number and the value of $|R_H|$.

Next let us discuss the frequency dependence of $\sigma_2/(\sigma_1\omega)$ in the c direction. As seen in Fig. 4, higher frequency data show smaller values along the c axis. This result is related to the fact that in this direction the surface resistance R_s at 14 GHz is greater than that at 9.6 GHz in the low temperature region [Fig. 1(a)]. This situation is similar to the millimeter-wave report of $CePd_3$ polycrystal by Beyermann *et al.*[11] They claimed that it originated from high-energy interband contribution to σ_2.[11,12] However, this explanation requires a large dielectric constant.[27] Thus, it is not clear whether such a high-energy contribution affects the microwave (low-frequency) conductivity in CeNiSn. Another possible explanation can be derived from the consideration of the frequency dependence of the scattering time. When the Drude model is generalized to allow the free carriers to have frequency-dependent scattering rate,[28,29] the observed increase in the c-axis surface resistance can be explained by

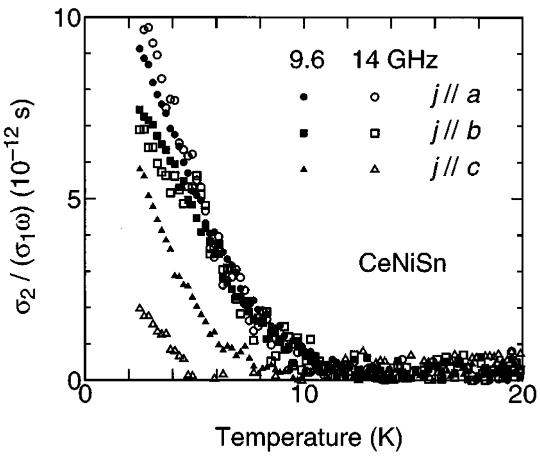

FIG. 4. Temperature dependence of $\sigma_2/(\sigma_1\omega)$ at 9.6 and 14 GHz along the a, b, and c directions in the CeNiSn single crystal. In the single-band Drude model [Eq. (3)], the vertical axis corresponds to the scattering time τ of the carriers [see Eq. (4)].

the increase in the scattering rate with increasing frequency. Such an anisotropy of $\tau(\omega,T)$ can be a reflection of the anisotropy of the coherence gap.[8] Quantitative analysis including higher-frequency measurements may help to elucidate the frequency-dependent and anisotropic scattering mechanism in this compound.

IV. SUMMARY

In summary, we have measured the temperature dependence of the anisotropic surface impedance $Z_s = R_s + iX_s$ of a CeNiSn single crystal at 9.6 and 14 GHz. At low temperatures, we found that the Hagen-Rubens relation ($R_s = X_s$) breaks down even in the microwave frequency region. The temperature dependence of complex conductivity was determined from the surface impedance data without using the Kramers-Kronig transformations. The real part σ_1 is much lower than the dc conductivity, and the imaginary part increases drastically at low temperatures. These results indicate that the scattering rate of the quasiparticles is rapidly reduced below 10 K in this system. The gap formation in this system reduces both the number and scattering rate of the carriers, which manifests itself as the metallic temperature dependence of the resistivity in clean samples.

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[1] T. Takabatake, Y. Nakazawa, and M. Ishikawa, Jpn. J. Appl. Phys. Suppl. **26**, 547 (1987).

[2] T. Takabatake, F. Teshima, H. Fujii, S. Nishigori, T. Suzuki, T. Fujita, Y. Yamaguchi, J. Sakurai, and D. Jaccard, Phys. Rev. B **41**, 9607 (1990).

[3] K. Nakamura, Y. Kitaoka, K. Asayama, T. Takabatake, H. Tanaka, and H. Fujii, J. Phys. Soc. Jpn. **63**, 433 (1994).

[4] T. Ekino, T. Takabatake, H. Tanaka, and H. Fujii, Phys. Rev. Lett. **75**, 4262 (1995).

[5] G. Nakamoto, T. Takabatake, Y. Bando, H. Fujii, K. Izawa, T. Suzuki, T. Fujita, A. Minami, I. Oguro, L. T. Tai, and A. A. Menovsky, Physica B **206&207**, 840 (1995).

[6] T. Takabatake, G. Nakamoto, T. Yoshino, H. Fujii, K. Izawa, S. Nishigori, H. Goshima, T. Suzuki, T. Fujita, K. Maezawa, T.

Hiraoka, Y. Okayama, I. Oguro, A. A. Menovsky, K. Neumaier, A. Brückl, and K. Andres, Physica B **223&224**, 413 (1996).
[7] Y. Inada, H. Azuma, R. Settai, D. Aoki, Y. Õnuki, K. Kobayashi, T. Takabatake, G. Nakamoto, H. Fujii, and K. Maezawa, J. Phys. Soc. Jpn. **65**, 1158 (1996).
[8] T. E. Mason, G. Aeppli, A. P. Ramirez, K. N. Clausen, C. Broholm, N. Stücheli, E. Bucher, and T. T. M. Palstra, Phys. Rev. Lett. **69**, 490 (1992).
[9] T. J. Sato, H. Kadowaki, H. Yoshizawa, T. Ekino, T. Takabatake, H. Fujii, L. P. Regnault, and Y. Ishikawa, J. Phys.: Condens. Matter **7**, 8009 (1995).
[10] T. Hiraoka, E. Kinoshita, T. Takabatake, H. Tanaka, and H. Fujii, Physica B **199&200**, 440 (1994).
[11] W. P. Beyermann, G. Grüner, Y. Dalicheouch, and M. B. Maple, Phys. Rev. Lett. **60**, 216 (1988); Phys. Rev. B **37**, 10 353 (1988).
[12] A. M. Awasthi, W. P. Beyermann, J. P. Carini, and G. Grüner, Phys. Rev. B **39**, 2377 (1989).
[13] T. Shibauchi, A. Maeda, H. Kitano, T. Honda, and K. Uchinokura, Physica C **203**, 315 (1992).
[14] D. A. Bonn, R. Liang, T. M. Riseman, D. J. Baar, D. C. Morgan, K. Zhang, P. Dosanjh, T. L. Duty, A. MacFarlane, G. D. Morris, J. H. Brewer, W. N. Hardy, C. Kallin, and A. J. Berlinsky, Phys. Rev. B **47**, 11 314 (1993).
[15] T. Shibauchi, A. Maeda, H. Kitano, T. Honda, and K. Uchinokura, *Proceedings of the Fifth International Symposium on Superconductivity*, edited by Y. Bando and H. Yamanouchi (Springer-Verlag, Tokyo, 1993), p. 175.
[16] S. Sridhar and W. L. Kennedy, Rev. Sci. Instrum. **59**, 531 (1988).
[17] The minimum value of skin depth is about 4 μm, whereas the mean free path at the lowest temperatures is estimated as 0.06 μm by using a simple relation $l = v_F \tau = \hbar \tau (3\pi^2 n)^{1/3}/m^*$. Therefore, the normal skin effect condition ($\delta_{cl} \gg l$) holds in the whole temperature range.
[18] T. Shibauchi, H. Kitano, K. Uchinokura, A. Maeda, T. Kimura, and K. Kishio, Phys. Rev. Lett. **72**, 2263 (1994).
[19] H. Kitano, T. Shibauchi, K. Uchinokura, A. Maeda, H. Asaoka, and H. Takei, Phys. Rev. B **51**, 1401 (1995).
[20] J. Mao, D. H. Wu, J. L. Peng, R. L. Greene, and S. M. Anlage, Phys. Rev. B **51**, 3316 (1995).
[21] K. Nakamura, Y. Kitaoka, K. Asayama, T. Takabatake, G. Nakamoto, H. Tanaka, and H. Fujii, Phys. Rev. B **53**, 6385 (1996).
[22] T. Shibauchi, H. Kitano, A. Maeda, H. Asaoka, H. Takei, I. Shigaki, T. Kimura, K. Kishio, K. Izumi, T. Suzuki, and K. Uchinokura, J. Phys. Soc. Jpn. **65**, 3266 (1996).
[23] The dc conductivity along the b and c axes also shows a maximum, which is different from high-T_c superconductors. In superconductors $\sigma_1(\omega)$ has the delta function at $\omega=0$, while in ordinary conductors $\sigma_1(\omega)$ at low frequency can be extrapolated to the dc conductivity by the Drude model.
[24] Y. Ishikawa, K. Mori, Y. Ogiso, K. Okabe, and K. Sato, J. Phys. Soc. Jpn. **60**, 2514 (1992); M. Sera, N. Kobayashi, T. Yoshino, K. Kobayashi, T. Takabatake, G. Nakamoto, and H. Fujii, Phys. Rev. B **55**, 6421 (1997).
[25] C. Uher and A. B. Kaiser, Phys. Rev. B **36**, 5680 (1987).
[26] H. Ikeda and K. Miyake, J. Phys. Soc. Jpn. **65**, 1769 (1996).
[27] P. E. Sulewski and A. J. Sievers, Phys. Rev. Lett. **63**, 2000 (1989).
[28] P. E. Sulewski, A. J. Sievers, M. B. Maple, M. S. Torikachvili, J. L. Smith, and Z. Fisk, Phys. Rev. B **38**, 5338 (1988).
[29] B. C. Webb, A. J. Sievers, and T. Mihalisin, Phys. Rev. Lett. **57**, 1951 (1986).