Non-Fermi liquid behavior and scaling of low frequency suppression in optical conductivity spectra of CaRuO$_3$

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Optical conductivity spectra $\sigma_1(\omega)$ of paramagnetic CaRuO$_3$ are investigated at various temperatures. At $T = 10$ K, it shows a non-Fermi liquid behavior of $\sigma_1(\omega) \sim 1/\omega^{\frac{3}{2}}$, similar to the case of a ferromagnet SrRuO$_3$. As the temperature ($T$) is increased, on the other hand, $\sigma_1(\omega)$ in the low frequency region is progressively suppressed, deviating from the $1/\omega^{\frac{3}{2}}$-dependence. Interestingly, the suppression of $\sigma_1(\omega)$ is found to scale with $\omega/T$ at all temperatures. The origin of the $\omega/T$ scaling behavior coupled with the non-Fermi liquid behavior is discussed.

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The Fermi liquid model has provided a fundamental concept in understanding metals. However, in some strongly correlated systems, non-Fermi liquid (NFL) behaviors have been often observed, where the Fermi liquid picture fails. In the normal state of high $T_c$ superconductors (HTS), evidences of NFL behaviors were reported in many experiments involving photoemission, transport, and optical measurements. In particular, their optical conductivity spectra $\sigma_1(\omega)$ show $1/\omega$-dependence in contrast to the usual Drude form of $\sim 1/\omega^2$, and their scattering rates show linear temperature- ($T$-) and $\omega$-dependences up to the mid-infrared (IR) region. Such unusual behaviors have been explained in terms of the marginal Fermi liquid.

Recently, perovskite ruthenates have attracted much attention as another class of materials exhibiting NFL behavior. The ruthenates belong to 4$d$ transition metal oxides, and the electron correlation effects are believed to play a crucial role in determining their physical properties. Together with their intriguing transport properties, the optical spectra show a distinct NFL behavior. For an itinerant ferromagnet SrRuO$_3$ (a ferromagnetic transition temperature $T_c = 165$ K), Kostic et al. reported that its $\sigma_1(\omega)$ at low $T$ follow a $1/\omega^{\frac{3}{2}}$-dependence, indicating a NFL behavior stronger than that of HTS. Recently, Dodge et al. fitted the $\sigma_1(\omega)$ of SrRuO$_3$ with $\sigma_1(\omega) \sim (\tau^{-1} - i\omega)^{-\alpha}$ with $\alpha \sim 0.4$ down to a very low energy region of $\sim 0.001$ eV. Although the NFL behavior in the ruthenate has been widely accepted, its origin is not clearly understood. In addition, an unusual suppression of $\sigma_1(\omega)$ in the low energy region occurs in the paramagnetic (PM) state, but is absent in the ferromagnetic (FM) state. However, this intriguing phenomenon, which might be coupled with a NFL behavior, has not been addressed properly.

While CaRuO$_3$ has the electronic structure similar to that of SrRuO$_3$, this material does not show any magnetic ordering down to a very low $T$. CaRuO$_3$ can provide a relatively wide $T$ window for investigating the interesting PM state as well as another example for understanding the NFL behavior. In this paper, we investigated the electrodynamic responses of CaRuO$_3$. It was found that $\sigma_1(\omega)$ at 10 K follows $\sim 1/\omega^{\frac{3}{2}}$, indicating a NFL behavior. With increasing $T$, the suppression of $\sigma_1(\omega)$ near $\omega \approx 0$ develops below the characteristic energy $\omega_c$, which corresponds to a peak structure in $\sigma_1(\omega)$ and shifts to higher frequencies as $T$ increases. It is remarkable that the low frequency optical spectra in a function of $\sigma_1(\omega)/\omega^{\frac{3}{2}}$ show a $\omega/T$ scaling behavior in a very wide $T$ range. While there have been similar scaling behavior reported in some physical properties of other NFL systems, the $\omega/T$ scaling behavior in ruthenates is the first observation in optical spectra. This scaling indicates that the only characteristic energy scale should be set by $T$ in the PM state of the perovskite ruthenates.
Several CaRuO$_3$ epitaxial films on (100) SrTiO$_3$ substrates were fabricated using 90° off-axis sputtering techniques. Their thicknesses were about 5000 Å. To obtain high crystalline quality films with little strain effect, we used vicinal substrates with large miscut angles (4° and 7°). The dc resistivity $\rho (T)$ was measured up to 500 K using the standard four probe method. Figure 1 shows the $\rho (T)$ curve of a film, which is nearly the same as that of a bulk single crystal, including a crossover near around 50 K below which the $T$-dependence changes from $T^2$ to $T^\frac{3}{2}$. The 300 K resistivity value of the film is $\sim 270 \mu$Ωcm, comparable to that of the bulk single crystal value of resistivity $\sim 200 \mu$Ωcm. The resistivity ratio $\rho (300 \text{K}) / \rho (10 \text{K})$ is about 9, indicating the high quality of our film. It is interesting that the $\rho (T)$ in the PM state of the perovskite ruthenates follows a $T^\frac{3}{2}$-dependence. The $\rho (T)$ of CaRuO$_3$ increases continuously up to 500 K with no saturation, and $\rho (T) \sim T^\frac{3}{2}$ above 50 K. Note that, as displayed in Fig. 1, the reported $\rho$ values of a single crystal SrRuO$_3$ in the PM state also show the $T^\frac{3}{2}$-dependence. The $T^\frac{3}{2}$-dependence of $\rho (T)$ in the perovskite ruthenates is another anomalous feature, distinguished from the linear $T$-dependence of $\rho (T)$ in the normal state of HTS.

Near normal incident reflectivity spectra $R(\omega)$ were measured in a wide photon energy region of 5 meV $\sim$ 30 eV. The Kramers-Kronig (K-K) analysis was used to calculate $\sigma_1 (\omega)$ from the measured $R(\omega)$. For K-K transformation, $R(\omega)$ in the low frequency region were extrapolated with the Hagen-Rubens relation. $T$-dependent $R(\omega)$ were measured in a photon energy region below 6 eV. Above 6 eV, the room temperature $R(\omega)$ was used for high frequency extrapolation. The overall features of the measured $R(\omega)$ were similar to those in SrRuO$_3$ reported by Kostic et al. The calculated $\sigma_1 (\omega)$ from the K-K analysis agreed with the experimental $\sigma_1 (\omega)$ independently obtained by spectroscopic ellipsometry in the visible region, which demonstrates the validity of our K-K analysis. A high frequency region of $\sigma_1 (\omega)$ in CaRuO$_3$ were described in our published paper. In the paper, we focus on the far-IR region.

Figure 2 shows $T$-dependent $\sigma_1 (\omega)$ in the far-IR region. The peak at $\sim 570 \text{cm}^{-1}$ is due to a transverse optic phonon mode, whose $T$-dependence is rather weak. Interestingly, $\sigma_1 (\omega)$ at 10 K shows a clear NFL behavior, deviating from that of conventional metals. As shown in the inset of Fig. 2, $\sigma_1 (\omega)$ at 10 K is proportional to $1/\omega^\frac{3}{2}$, which is much slower than the frequency dependence of a Fermi liquid of $1/\omega^2$. Even at a higher $T$, the $1/\omega^\frac{3}{2}$-dependence in $\sigma_1 (\omega)$ is retained in the high frequency region, which might be correlated with the $T^\frac{3}{2}$-dependence of $\rho (T)$ at high temperatures. A similar NFL behavior was also observed in SrRuO$_3$. It is interesting that the $1/\omega^\frac{3}{2}$-dependence in $\sigma_1 (\omega)$ can be observed in perovskite ruthenates with different magnetic ground states.
where the dc conductivity value is rather high by $\sim 7000\ \Omega^{-1}\text{cm}^{-1}$. It is noted that in the case of SrRuO$_3$, the low frequency suppression occurs only in its PM state, not in its FM state. Together with the $T^2$-dependence of $\rho(T)$, the low-energy suppression of $\sigma_1(\omega)$ can be regarded as a generic feature of the PM state of the perovskite ruthenates.

Note that the low-energy suppression of $\sigma_1(\omega)$ shows an interesting $T$-dependent evolution. As $T$ increases, the suppression feature becomes clear and $\omega_c$ shifts to a higher frequency linearly with $T$. It is evident that the peak structure does not arise from electronic transitions or disorder effects. The values of $\omega_c$ are comparable with a thermal energy, $k_B T$. This energy scale is too low for a typical interband transition. A similar suppression in $\sigma_1(\omega)$ near $\omega \approx 0$ has been often observed in highly disordered systems, but their characteristic energy scale is expected to decrease with increasing $T$ which is the opposite to our case. Therefore, a thermal energy scale of a pseudogap-like feature observed in CaRuO$_3$ is quite unique.

The pseudogap-like feature could be closely related to nearly ferromagnetic characteristics of CaRuO$_3$. Several experimental and theoretical evidences suggest that CaRuO$_3$ should be nearly ferromagnetic. A strong FM fluctuation was also observed in the PM state of SrRuO$_3$. Especially, local density functional calculations on (Ca,Sr)RuO$_3$ showed that the lattice distortions associated with different ionic sizes are crucial in determining the magnetic properties. Further, phonon anomalies at $T_c$ were observed in SrRuO$_3$, indicating the strong spin-lattice interaction. These imply that the lattice degree of freedom is strongly coupled to the magnetic ordering so that the excitation of a relevant phonon mode can be responsible for the local magnetic fluctuation in the nearly FM system. From the spin fluctuation theory of nearly ferromagnetic materials, it is known that the mean-square amplitude of spin-fluctuation increases linearly in proportion to $T$. Thus, one may expect the low-energy quasiparticle excitations to be strongly renormalized by the thermally induced spin-fluctuations with spin-lattice coupling, where such $T$-dependent renormalization might be relevant to the suppression of $\sigma_1(\omega)$ in the low frequency region. It is noted that the pseudogap-like feature in $\sigma_1(\omega)$ of CaRuO$_3$ and its proximity to the FM instability is quite analogous to the situation in HTS, which is close to the antiferromagnetic instability.

Now, we show that from the systematic $T$-dependent evolution of the low frequency suppression in $\sigma_1(\omega)$, an interesting $\omega/T$ scaling behavior can occur in the perovskite ruthenates. The low frequency suppression in $\sigma_1(\omega)$ can be expressed as a deviation from the $1/\omega^2$ dependence. As shown in the inset of Fig. 2, the deviation region of $\sigma_1(\omega)$ from $1/\omega^2$-dependence becomes wider with increasing $T$, consistent with the shift of $\omega_c$ to higher frequency. To check the possibility of scaling behavior, we plotted $\sigma_1(\omega)/a\omega^{-2}$ vs. $\omega/T$. With the value of the coefficient $a$ adopted for the scaling, the $\sigma_1(\omega)$ at 10 K was reproduced. As shown in Fig. 3, all of the normalized conductivity spectra collapse onto a single line. It is noted that this scaling behavior persists up to a rather high temperature, 500 K. The $\omega/T$ scaling behavior means that the $T$-dependent suppression behavior could be determined only by $T$, indicating

$$\sigma_1(\omega) \sim 1/\omega^2 \cdot Z(\omega/T), \quad (1)$$

or

$$\sigma_1(\omega)T^{\frac{\beta}{2}} \sim (T/\omega)^{\frac{2}{2}} \cdot Z(\omega/T), \quad (2)$$

The scaling function $Z(\omega/T)$ is fitted quite well with $Z(\omega/T) = \tanh(\beta \omega/T)$, with $\beta = 1.6$. Clearly, Eq. (1) and (2) are closely related to the characteristic properties in the PM states, such as $\rho(T) \sim T^\frac{1}{2}$ and $\sigma_1(\omega) \sim 1/\omega^2$ at high frequencies. We also plotted the SrRuO$_3$ $\sigma_1(\omega)$ data in the PM region (i.e., at $185\ K$, $225\ K$, and $250\ K$) reported by Kostic et al. Interestingly, the normalized spectra of SrRuO$_3$ fall on the scaling curve. This indicates that the scaling function shown in Fig. 3 could be applied to other perovskite ruthenates.

![Fig. 3. T-dependent $\sigma_1(\omega)/a\omega^{-2}$ with $\omega/T$ as the abscissa. The open circle, open trigonal, and cross symbols are for the 185 K, 225 K, and 250 K spectra of SrRuO$_3$, respectively, quoted from Ref. [7]. For SrRuO$_3$, the value of $a$ was adopted to reproduce the 40 K $\sigma_1(\omega)$. The solid line represents $Z(\omega/T) = \tanh(1.6\omega/T)$.](image-url)
fluctuation associated with the zero temperature phase transition. Similar to other NFL systems, our $\omega/T$ scaling behavior in $\sigma_1(\omega)$ may suggest a quantum critical point between the ferromagnetic and paramagnetic phases in the perovskite ruthenates: The ferromagnetic transition temperature $T_c$ is decreased as $x$ is increased in Sr$_{1-x}$Ca$_x$RuO$_3$ and is completely suppressed in CaRuO$_3$, and a quantum critical point is expected at an appropriate value of $x = x_c$. We note that, consistent with the magnetic quantum phase transition, the previous low $T$ transport measurements hinted a phase transition from a Fermi liquid behavior for SrRuO$_3$ to a NFL behavior for CaRuO$_3$. Motivated by our observation, understanding the origin of the NFL behavior associated with the $\omega/T$ scaling in $\sigma_1(\omega)$ and its possible relation with the quantum criticality is a challenging issue in the future study of perovskite ruthenates.

In summary, the optical spectra of the nearly ferromagnetic CaRuO$_3$ shows non-Fermi liquid behavior and a scaling in the low frequency suppression. Its $\sigma_1(\omega)$ follows the $1/\omega^{2}$-dependence, similar to the case of SrRuO$_3$. From the $T$-dependent evolution of the low frequency suppression, it is observed that the $\sigma_1(\omega)$ normalized by the $1/\omega^{2}$ can be scaled with $\omega/T$ at all temperatures, indicating that only characteristic energy scale is determined by $T$. The $\omega/T$ scaling coupled with the non-Fermi liquid behavior is expected to provide further insights into understanding the unusual electrodynamics of the ruthenates.

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1 D. Pines and P. Nozieres, *The Theory of Quantum Liquid, Vol 1: Normal Fermi Liquids* (Addison-Wesley, Reading, MA, 1987).
2 J. Orenstein and A. J. Millis, Science 288, 468 (2000).
3 C. M. Varma, P. B. Littlewood, S. Schmitt-Rink, E. Abrahams, and A. E. Ruckenstein, Phys. Rev. Lett. 63, 1996 (1989).
4 J. S. Lee, Y. S. Lee, T. W. Hoh, K. Char, J. Park, S. J. Oh, J. H. Park, C. B. Eom, T. Takeda, and R. Kanno, Phys. Rev. B 64, 245107 (2001), and the references therein.
5 P. B. Allen, H. Berger, O. Chauvet, L. Forro, T. Jarlborg, A. Junod, B. Revaz, and G. Santi, Phys. Rev. B 53, 4393 (1996).
6 L. Klein, L. Antognazza, T. H. Geballe, M. R. Beasley, and A. Kapitulnik, Phys. Rev. B 60, 1448 (1999).
7 P. Kostic, Y. Okada, N. C. Collins, Z. Schlesinger, J. W. Reiner, L. Klein, A. Kapitulnik, T. H. Geballe, and M. R. Beasley, Phys. Rev. Lett. 81, 2498 (1998).
8 J. S. Dodge, C. P. Weber, J. Corson, J. Orenstein, Z. Schlesinger, J. W. Reiner, and M. R. Beasley, Phys. Rev. Lett. 85, 4932 (2000).
9 G. Cao, S. McCall, M. Shepard, J. E. Crow, and R. P. Guertin, Phys. Rev. B 56, 321 (1997).
10 M. C. Aronson, R. Osborn, R. A. Robinson, J. W. Lynn, R. Chau, C. L. Seaman, and M. B. Maple, Phys. Rev. Lett. 75, 725 (1995).
11 A. Schröder, G. Apell, E. Bucher, R. Ramazashvili, and P. Coleman, Phys. Rev. Lett. 80, 5623 (1998). T. Valla, A. V. Fedorov, P. D. Johnson, B. O. Wells, S. L. Hulbert, Q. Li, G. D. Gu, and N. Koshizuka, Science 285, 2110 (1999).
12 C. B. Eom, R. J. Cava, R. M. Fleming, J. M. Phillips, R. B. van Dover, J. H. Marshall, J. W. Hsu, J. J. Krajewski, and W. F. Peck, Jr., Science 258, 1766 (1992). R. A. Rao, Q. Gan, C. B. Eom, R. J. Cava, Y. Suzuki, J. J. Krajewski, S. C. Gauze, Pohli, and M. Lee, Appl. Phys. Lett. 70, 3035 (1997).
13 Y. S. Lee, J. S. Lee, K. W. Kim, T. W. Noh, J. Yu, Y. Bang, M. K. Lee, and C. B. Eom, Phys. Rev. B 64, 165109 (2001).
14 From the extended Drude model analysis, it is found that the frequency-dependent scattering rate at 10 K follows the $\omega^{2}$-dependence with the correlation to the $1/\omega^{2}$-dependence in $\sigma_1(\omega)$.
15 D. N. Basov, B. Dabrowski, and T. Timusk, Phys. Rev. Lett. 81, 2132 (1998). A. Gold, S. J. Allen, B. A. Wilson, and D. C. Tsui, Phys. Rev. B 25, 3519 (1982).
16 K. Yoshimura, T. Imai, T. Kiyama, K. R. Thurbler, A. W. Hunt, and K. Kosuge, Phys. Rev. Lett. 83, 4397 (1999).
17 T. He and R. J. Cava, Phys. Rev. B 63, 172403 (2001).
18 I. I. Mazin and D. J. Singh, Phys. Rev. B 56, 2556 (1997).
19 M. N. Iliev, A. P. Litvinchuk, H.-G. Lee, C. L. Chen, M. L. Dezanetti, C. W. Chu, V. G. Ivanov, M. V. Abrishev, and V. N. Popov, Phys. Rev. B 59, 364 (1999).
20 T. Moriya, *Spin Fluctuations in Itinerant Electron Magnetism* (Springer-Verlag, New York, 1985).
21 D. Manske, I. Eremin, and K. H. Bennemann, Phys. Rev. Lett. 87, 177005 (2001).
22 It is not clear that even the 10 K $\sigma_1(\omega)$ follows the scaling behavior. If so, this might be observed below 20 cm$^{-1}$, where is beyond the experimental region.
23 S. Sachdev, *Quantum Phase Transition* (Cambridge Univ. Press, Cambridge, 1999).
24 F. Fukunaga and N. Tsuda, J. Phy. Soc. Jpn 63, 3798 (1994).
25 L. Capogna, A. P. Mackenzie, R. S. Perry, S. A. Grigera, L. M. Galvin, P. Raychaudhuri, A. J. Schofield, C. S. Alexander, G. Cao, S. R. Jullian, and Y. Maeno, Phys. Rev. Lett. 88, 76602 (2002).