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Non-Fermi liquid behavior and scaling of the low-frequency suppression in the optical conductivity spectra of $CaRuO_3$

Y. S. Lee and Jaejun Yu
School of Physics and Center for Strongly Correlated Materials Research, Seoul National University, Seoul 151-747, Korea

J. S. Lee and T. W. Noh
School of Physics and Research Center for Oxide Electronics, Seoul National University, Seoul 151-747, Korea

T.-H. Gimm and Han-Yong Choi
School of Physics and Institute for Basic Science Research, Sung Kyun Kwan University, Suwon 440-746, Korea

C. B. Eom
Department of Material Science and Engineering, University of Wisconsin-Madison, Madison, Wisconsin 53706
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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 $\sigma_1(\omega)\sim 1/\omega^{1/2}$, 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 $1/\omega^{1/2}$ dependence. Interestingly, the suppression of $\sigma_1(\omega)$ is found to scale with ω/T at all temperatures. The origin of the ω/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.[1] 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.[2] 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 ω dependences up to the mid-IR region. Such unusual behaviors have been explained in terms of the marginal Fermi liquid.[3]

Recently, perovskite ruthenates have attracted much attention as another class of materials exhibiting NFL behavior. The ruthenates belong to $4d$ transition-metal oxides, and the electron correlation effects are believed to play a crucial role in determining their physical properties.[4] Together with their intriguing transport properties,[5,6] the optical spectra show a distinct NFL behavior.[7,8] For an itinerant ferromagnet $SrRuO_3$ (a ferromagnetic transition temperature $T_c=165$ K), Kostic *et al.*[7] reported that its $\sigma_1(\omega)$ at low T follow a $1/\omega^{1/2}$ dependence, indicating a NFL behavior stronger than that of HTS. Recently, Dodge *et al.*[8] fitted the $\sigma_1(\omega)$ of $SrRuO_3$ with $\sigma_\alpha(\omega)\sim(\tau^{-1}-i\omega)^{-\alpha}$ with $\alpha\sim 0.4$ down to a very low-energy region of ~ 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.[8] 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.[9] $CaRuO_3$ 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 $\sim 1/\omega^{1/2}$, indicating a NFL behavior. With increasing T, the suppression of $\sigma_1(\omega)$ near $\omega\simeq 0$ develops below the characteristic energy ω_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 $\sigma_1(\omega)/\omega^{-1/2}$ show a ω/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,[10,11] the ω/T scaling behavior in ruthenates is an 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.[12] 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 $T^{1/2}$ to $T^{3/2}$.[9] The 300-K resistivity value of the film is ~ 270 $\mu\Omega$ cm, comparable to that of the bulk single-crystal value of resistivity ~ 200 $\mu\Omega$ cm.[9] The resistivity ratio $\rho(300\text{ K})/\rho(10\text{ K})$ is about 9, indicating the high quality of our film. It is interest-

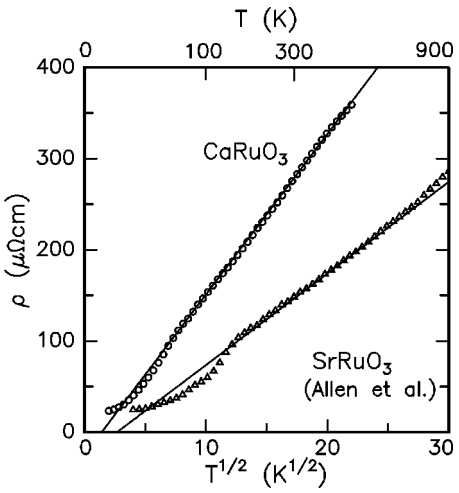

FIG. 1. T-dependent $\rho(T)$ curves of a $CaRuO_3$ film (open circle) and a $SrRuO_3$ single crystal (open triangle). The data of $SrRuO_3$ are quoted from Ref. 5.

ing that the $\rho(T)$ in the PM state of the perovskite ruthenates follows a $T^{1/2}$ dependence. The $\rho(T)$ of $CaRuO_3$ increases continuously up to 500 K with no saturation, and $\rho(T) \sim T^{1/2}$ above 50 K.[6] Note that, as displayed in Fig. 1, the reported ρ values of a single crystal $SrRuO_3$ in the PM state also show the $T^{1/2}$ dependence.[5] The $T^{1/2}$ 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–30 eV. The Kramers-Kronig (KK) analysis was used to calculate $\sigma_1(\omega)$ from the measured $R(\omega)$. For KK 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.*[7] The calculated $\sigma_1(\omega)$ from the KK analysis agreed with the experimental $\sigma_1(\omega)$ independently obtained by spectroscopic ellipsometry in the visible region, which demonstrates the validity of our KK analysis.[13] A high-frequency region of $\sigma_1(\omega)$ in $CaRuO_3$ were described in our published paper.[4] In the paper, we focus on the far-IR region.

Figure 2 shows the T-dependent $\sigma_1(\omega)$ in the far-IR region. The peak at $\sim 570\ \mathrm{cm}^{-1}$ 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 $1/\omega^{1/2}$,[14] which is much slower than the frequency dependence of a Fermi liquid of $1/\omega^2$. Even at a higher T, the $1/\omega^{1/2}$ dependence in $\sigma_1(\omega)$ is retained in the high-frequency region, which might be correlated with the $T^{1/2}$ dependence of $\rho(T)$ at high temperatures. A similar NFL behavior was also observed in $SrRuO_3$.[7] It is interesting that the $1/\omega^{1/2}$ dependence in $\sigma_1(\omega)$ can be observed in perovskite ruthenates with different magnetic ground states.

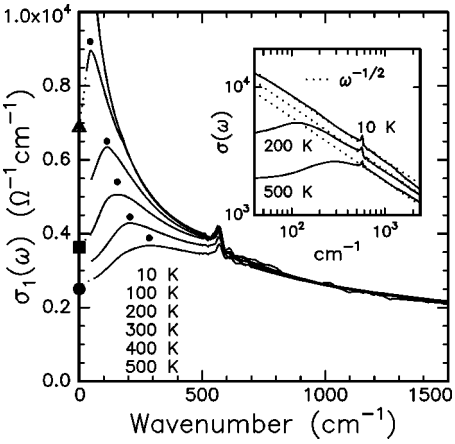

FIG. 2. T-dependenct $\sigma_1(\omega)$ of $CaRuO_3$ below 1500 cm^{-1}. The solid circle symbols represent ω_c. The solid triangle, the solid square, and the solid circle symbols on the y axis represent the dc value of $\sigma(T)$ at 100, 300, and 500 K, respectively. The dotted lines are $\sigma_1(\omega)$ obtained from the Hagen-Rubens extrapolations. In the inset, $\sigma_1(\omega)$ at 10, 200, 500 K follow $1/\omega^{1/2}$ above ω_c. For clarity, the $\sigma_1(\omega)$ at 200 K and 500 K are multiplied by a factor of 0.85 and 0.7, respectively. The dotted lines are guidelines for $1/\omega^{1/2}$ dependence.

The suppression in $\sigma_1(\omega)$ is observed near $\omega \simeq 0$ at higher T. As ω decreases from the high-frequency side, $\sigma_1(\omega)$ increases initially but decrease below the peak frequency ω_c, approaching smoothly to the measured dc conductivity values. This feature is clear even at 100 K, where the dc conductivity value is rather high by $\sim 7000\ \Omega^{-1}\,\mathrm{cm}^{-1}$. 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.[8] Together with the $T^{1/2}$ dependence of $\rho(T)$, the low-energy suppression in $\sigma_1(\omega)$ 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 ω_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 ω_c are comparable with a thermal energy, $k_B T$. This energy scale is too low for a typical interband transition.[4] A similar suppression in $\sigma_1(\omega)$ near $\omega \simeq 0$ has been often observed in highly disordered systems, but their characteristic energy scale is expected to decrease with increasing T,[15] which is 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.[16–18] A strong FM fluctuation was also observed in the PM state of $SrRuO_3$.[16] 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.[18] Further, phonon anomalies at T_c were observed in $SrRuO_3$,

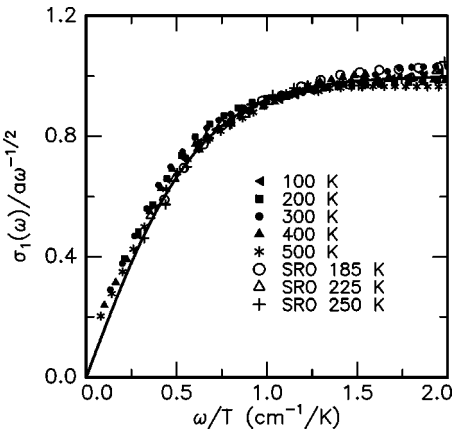

FIG. 3. T-dependent $\sigma_1(\omega)/a\omega^{-1/2}$ with ω/T as the abscissa. The open circle, open trigonal, and cross symbols are for the 185 K, 225 K, and 250 K spectra of $SrRuO_3$, respectively, quoted from Ref. 7. For $SrRuO_3$, the value of a was adopted to reproduce the 40 K $\sigma_1(\omega)$. The solid line represents $Z(\omega/T)=\tanh(1.6\omega/T)$.

indicating the strong spin-lattice interaction.[19] 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,[20] 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.[21]

Now, we show that from the systematic T-dependent evolution of the low-frequency suppression in $\sigma_1(\omega)$, an interesting ω/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 $1/\omega^{1/2}$ dependence. As shown in the inset of Fig. 2, the deviation region of $\sigma_1(\omega)$ from $1/\omega^{1/2}$ dependence becomes wider with increasing T, consistent with the shift of ω_c to higher frequency. To check the possibility of scaling behavior, we plotted $\sigma_1(\omega)/a\omega^{-1/2}$ vs ω/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.[22] It is noted that this scaling behavior persists up to a rather high temperature, 500 K. The ω/T scaling behavior means that the T-dependent suppression behavior could be determined only by T, indicating

$$\sigma_1(\omega)\sim 1/\omega^{1/2}Z(\omega/T), \tag{1}$$

or

$$\sigma_1(\omega)T^{1/2}\sim(T/\omega)^{1/2}Z(\omega/T). \tag{2}$$

The scaling function $Z(\omega/T)$ is fitted quite well with $Z(\omega/T)=\tanh(\beta\omega/T)$, with $\beta=1.6$. Clearly, Eqs. (1) and (2) are closely related to the characteristic properties in the PM states, such as $\rho(T)\sim T^{1/2}$ and $\sigma_1(\omega)\sim 1/\omega^{1/2}$ 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.*[7] 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.

While the ω/T scaling behavior in $\sigma_1(\omega)$ of the perovskite ruthenates is quite unique, it is noted that similar scale invariance has also been observed in some physical properties of other NFL systems, such as some f-electron compounds[10] and HTS.[11] The scaling behaviors in these NFL systems indicate that the only characteristic energy scale is determined by T, a possible origin of which was suggested to be the quantum critical fluctuation associated with the zero-temperature phase transition.[23] Similar to other NFL systems, our ω/T scaling behavior in $\sigma_1(\omega)$ may suggest a quantum critical point between the ferromagnetic and paramagnetic phases in the perovskite ruthnates: The ferromagnetic transition temperature T_c is decreased as x is increased in $Sr_{1-x}Ca_xRuO_3$ and is completely suppressed in $CaRuO_3$, and a quantum critical point is expected at an appropriate value of $x=x_c$.[9,24] 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 $CaRuO_3$.[24,25] Motivated by our observation, understanding the origin of the NFL behavior associated with the ω/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 $CaRuO_3$ show non-Fermi-liquid behavior and a scaling in the low-frequency suppression. Its $\sigma_1(\omega)$ follows the $1/\omega^{1/2}$-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 $1/\omega^{1/2}$ can be scaled with ω/T at all temperatures, indicating that only characteristic energy scale is determined by T. The ω/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* (Addison-Wesley, Reading, MA, 1987), Vol. 1.
[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. Noh, 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. Aeppli, 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.P. 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. Gausepohl, 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 $\omega^{1/2}$ dependence with the correlation to the $1/\omega^{1/2}$ 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. Thurber, 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. Dezaneti, C.W. Chu, V.G. Ivanov, M.V. Abrashev, and V.N. Popov, Phys. Rev. B **59**, 364 (1999).
[20] T. Morya, *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 at 10 K $\sigma_1(\omega)$ follows the scaling behavior. If so, this might be observed below 20 cm^{-1}, which is beyond the experimental region.
[23] S. Sachdev, *Quantum Phase Transition* (Cambridge University Press, Cambridge, England, 1999).
[24] F. Fukunaga and N. Tsuda, J. Phys. 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**, 076602 (2002).