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Enhanced paramagnetism of the 4*d* itinerant electrons in the rhodium oxide perovskite $SrRhO_3$

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Polycrystalline rhodium (IV) oxide perovskite $SrRhO_3$ was obtained by high-pressure synthesis techniques, followed by measurements of the magnetic susceptibility, electrical resistivity, and specific heat. The title compound has five 4*d* electrons per perovskite unit and shows Fermi-liquid behavior in its electrical resistivity. The magnetic susceptibility is large $[\chi(300\ \mathrm{K}) \sim 1.1 \times 10^{-3}$ emu/mol Rh$]$ and proportional to $1/T^2$ $(<380\ \mathrm{K})$, while there is no magnetic long-range order above 1.8 K. The specific heat measurements indicate a probable magnetic contribution below ~ 15 K, which is not predicted by the self-consistent renormalization theory of spin fluctuations for both antiferro- and ferromagnetic three-dimensional nearly ordered metals.

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I. INTRODUCTION

Since *p*-wave symmetrical pairing of electrons was proposed, driven mainly by ferromagnetic spin fluctuations, in the 1.5 K superconductor Sr_2RuO_4,[1] further superconducting phases have been expected in the vicinity of the 214 phase. This is probably due to substantial spin fluctuations found in neighboring compounds, including ferromagnetic $SrRuO_3$ (Refs. 2 and 3) and nearly ferromagnetic $CaRuO_3$ (Refs. 3 and 4) and $Sr_3Ru_2O_7$ (Ref. 5). Although intensive investigations have been applied to the ruthenium oxide systems, further "*p*-wave" superconducting phases have not been discovered thus far. The current experimental studies on ferromagnetically induced superconductivity, then, seem to be tied to a very local variety of materials. To ameliorate the stagnant situation, we have been exploring other correlated 4*d*-metal compounds, not only to find further superconducting materials in the ruthenium oxide system, but also to expand the variety of potential chemical systems for the spin-fluctuations-induced superconductors.

The rhodium oxide perovskite $SrRhO_3$ was recently found, and a pure polycrystalline sample was obtained by high-pressure synthesis techniques at 60 kbar and 1500 °C, followed by investigations of the magnetic susceptibility, electrical resistivity, and specific heat. The compound was fairly metallic and showed enhanced and thermally activated paramagnetism in the studied temperature range below 380 K. A qualitative fit of the Curie-Weiss (CW) law to the magnetic susceptibility data yielded a negative Weiss temperature of -361 K, if the analysis provided a correct sense of the magnetism. Neither superconductivity nor long-range magnetic order was found above 1.8 K. The magnetic data for $SrRhO_3$ appeared to be qualitatively similar to what was observed for the analogous ruthenium oxide metal $CaRuO_3$.[3,4] Since then, the self-consistent renormalization (SCR) theory of spin fluctuations for both antiferro- and ferromagnetic nearly ordered magnetic metals was tested on the observed electronic properties as was done for $CaRuO_3$.[3,4] As a result, all of the present data for $SrRhO_3$ do not meet the quantitative expectations as predicted by the theory.

II. EXPERIMENT

Variable composition precursors were prepared at Sr:Rh $=1:3$, 1:2, and 1:1 as follows. Mixtures of pure $SrCO_3$ (99.9%) and Rh (99.9%) powders were heated in oxygen at 1000 °C overnight, and then ground well and reheated in oxygen at 1200 °C for 2 days.[6] One and two moles of SrO_2 $(>99.9\%)$ were added to the 1:2 and 1:3 precursors per the formula, respectively, and 8 wt % of $KClO_4$ to the 1:1 precursor. Those were mixed well, and approximately 0.2 g of each were placed into Pt capsules. Those were heated at 60 kbar and 1500 °C for 1 h, then quenched to room temperature before releasing the pressure.[7] The quality of the finally obtained pellets was studied by powder x-ray-diffraction techniques in a regular manner. It was determined from the x-ray readings that the major phase was of perovskite type. The position and intensity distribution of the peaks for the phase were invariable among the patterns for every sample. The impurity level was 1% or less in every final production except KCl. The perovskite-type phase was denoted $SrRhO_3$, of which no records were found thus far in the literature.

Further structural characterization was made for the selected sample, which was prepared from the 1:2 precursor and SrO_2, by the x-ray Rietveld technique (Cu $K\alpha$) using the program RIETAN-2000.[8] A distorted perovskite structure model, $GdFeO_3$ type, was tested and found reasonable to describe the structure of $SrRhO_3$. The x-ray powder pattern and crystal structure are indicated in Fig. 1. The space group was *Pnma* (No. 62) and lattice parameters were $a=5.5394(2)$ Å, $b=7.8539(3)$ Å, and $c=5.5666(2)$ Å. The estimated positions for the atoms were Sr(0.0304(1),0.25,−0.0054(8)), Rh(0, 0, 0.5), O(1)(0.4990(23), 0.25, 0.0587(45)), and O(2)(0.2825(26), 0.0366(24), 0.7088(26)). During the refinement, the occupation factors, and the isotropic displacement parameters of the metals and oxygen were fixed at 1,

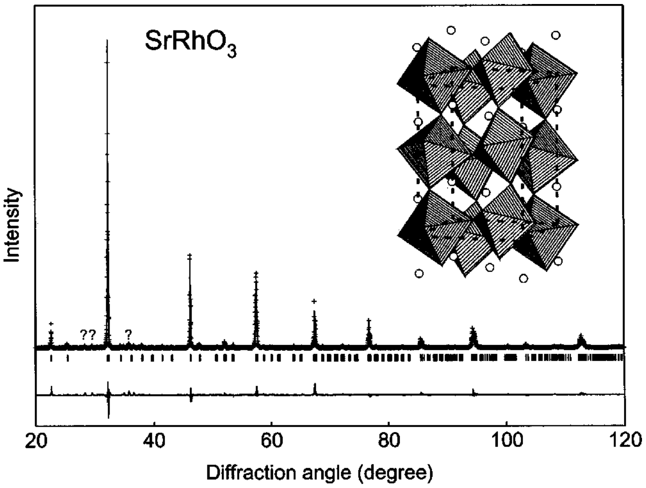

FIG. 1. The powder x-ray diffraction pattern (Cu$K\alpha$) for SrRhO$_3$. The vertical bars show the Bragg peak positions for SrRhO$_3$. The difference plot between the orthorhombic model pattern (solid lines) and the data (crosses) is shown below the bars. Unknown peaks are marked by "?." The crystal structure sketch with the orthorhombic unit cell (dotted lines) is shown as an inset. Open circles and polyhedra indicate Sr atoms and RhO$_6$ octahedra, respectively.

0.3, and 0.7, respectively. The final reliability factors and goodness of fit to the analysis were $R_{\mathrm{wp}}=20.9\%$, $R_{\mathrm{p}}=14.41\%$, $R_{\mathrm{R}}=18.57\%$, and $S=1.53$. Oxygen vacancies in the perovskite were quantitatively investigated in detail by thermogravimetric analysis and found to be insignificant.[9]

The same sample was again selected for characterization by magnetic, specific heat, and electrical resistivity measurements. The temperature dependence of magnetization was measured in a Quantum Design MPMS magnetometer. The specific heat and the electrical resistivity data were obtained in a Quantum Design PPMS apparatus. Those measurements were conducted between 1.8 and 400 K. The highest applied magnetic field was 70 kOe.

III. RESULTS AND DISCUSSIONS

The temperature dependence of the electrical resistivity of polycrystalline SrRhO$_3$ is shown in Fig. 2. The data were obtained by a standard four-terminal dc technique at a current of 5 mA on a piece of the sample pellet. The data clearly reveal the metallic nature of SrRhO$_3$; a metallic temperature dependence and ~ 1.3 mΩ cm at room temperature are typical for polycrystalline oxide metals. The low-temperature part (<50 K) is expanded and replotted as ρ vs T^2 (inset in Fig. 2). The observed linear dependence is indicative of Fermi-liquid behavior for SrRhO$_3$.[10] Subsequent fitting studies with standard resistivity expression for a Fermi liquid ($\rho=\rho_0+AT^2$) yielded $\rho_0=142$ $\mu\Omega$ cm and $A=0.062$ $\mu\Omega$ cm/K^2. The unusually large ρ_0 probably reflects contributions from extrinsic origins such as grain boundaries. The parameter ρ_0 was not constant among the sets of resistivity data for all of the present pellets (approximately two magnitudes larger for the pellet containing KCl), while the residual resistivity ratio ρ_{300}/ρ_0 remained almost constant (~ 9) among them. At the magnetic instability point or in the extreme vicinity of that point, the electrical resistivity is not expected to obey the famous T^2 law due to the influence of spin fluctuations; i.e., the $T^{3/2}$ and $T^{5/3}$ laws may be obeyed by antiferro- and ferromagnetically unstable three-dimensional (3D) metals, respectively.[11–13] A detailed analysis was preliminarily applied for the present resistivity data; however, non-Fermi-liquid behavior was not clearly seen. The magnetoresistivity at 1.8 K between -70 and 70 kOe was not observed and may be due to the polycrystalline nature of the sample. Additional studies using a single-crystal SrRhO$_3$, if it becomes available, could allow us to exclude extrinsic contributions and then might help to reveal the intrinsic nature of the electrical resistivity of SrRhO$_3$. Because the $4d$ band in SrRhO$_3$ is expected to be broad, as is the case for SrRuO$_3$, $4d$ electrons in the rhodium oxide should be itinerate by analogy.[2] The observed metallic conductivity is, hence, reflecting mainly the nature of unlocalized $4d$ electrons. The perovskite SrRhO$_3$ could be in a class of the itinerant $4d$-electron systems, such as (Sr,Ca)RuO$_3$.[2] Further investigations into the electronic transport of SrRhO$_3$, including band structure calculations, would be of interest.

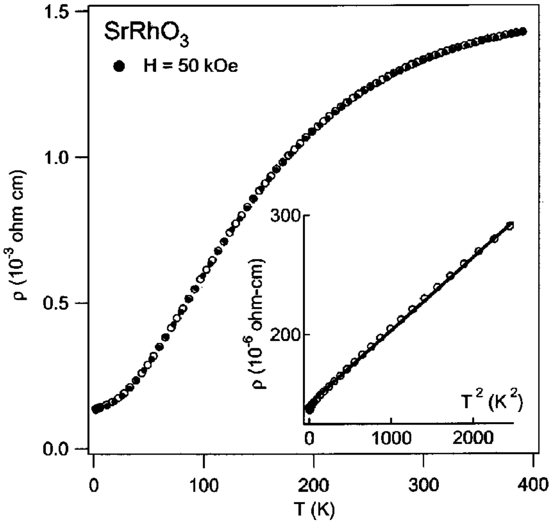

FIG. 2. The resistivity data for the polycrystalline SrRhO$_3$ measured between 1.8 and 390 K with and without a magnetic field of 50 kOe. The squared temperature plot for the data below 50 K is shown in the inset, indicating a linear dependence as shown by the solid line.

Magnetic data are summarized in Fig. 3. The magnetic susceptibility of SrRhO$_3$ obviously depends on temperature and is approximately 1.1×10^{-3} emu/mol Rh at room temperature, in contrast with the properties of the Pauli paramagnetic rhodium metal (approximately one magnitude smaller and almost temperature independent).[14] A steep rise in the χ vs T plot in low temperature at 10 kOe was observed, while it was significantly suppressed at 50 kOe. The M vs H curve at 2 K (inset in Fig. 3) indicates a subtle spontaneous magnetic moment ($\sim 0.001\mu_{\mathrm{B}}$ per Rh), suggesting SrRhO$_3$ has ordered magnetic moments. After subtraction of the major part, the $1/\chi_{\mathrm{upturn}}$ vs T plot results in a standard CW line with an insignificant level of Weiss tem-

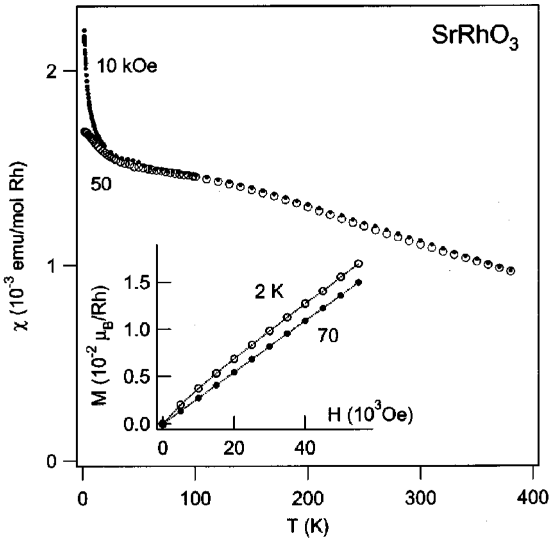

FIG. 3. The magnetic data for the polycrystalline $SrRhO_3$ measured between 1.8 and 400 K. Magnetic susceptibility vs temperature at 10 and 50 kOe are shown as solid and open circles, respectively, and the field dependence of magnetization at 2 and 70 K (inset).

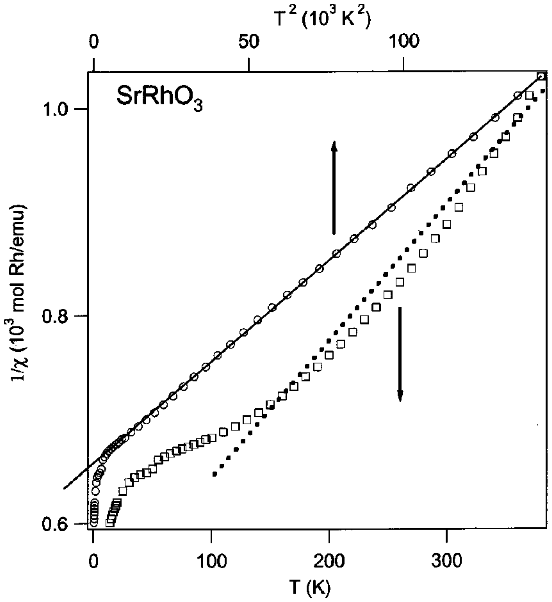

FIG. 4. Variable plotting of the reciprocal magnetic susceptibility of $SrRhO_3$. The contribution from the sample holder was carefully subtracted before the plotting. The dotted line represents preliminarily applied CW law to the $1/\chi$ vs T plot at $p_{\mathrm{eff}}=2.46\mu_{\mathrm{B}}$ and $\theta_{\mathrm{W}}=-361$ K. The $1/\chi$ vs T^2 plot shows a notably linear dependence as the solid line indicates.

perature, ~ -1.5 K.[15] It is therefore reasonable to conclude that the upturn results from a magnetic impurity origin rather than an ordered state of $SrRhO_3$. The slightly positive curvature of the M vs H curve at 2 K is probably due to superimposing a small amount of impurity component on the major part.

To further analyze the major part of the magnetic data for $SrRhO_3$, two plots of the reciprocal magnetic susceptibility were prepared in the forms of $1/\chi$ vs T and $1/\chi$ vs T^2 without any other manipulations except subtraction of sample holder contribution (Fig. 4). It is clearly seen in the temperature range that $1/\chi$ is proportional to T^2 rather than proportional to T as expected from the standard CW expression. Alternatively, the CW law with a temperature-independent term, i.e., $1/\chi = 1/[C/(T-\theta)+\chi_0]$ (C and θ are the Curie constant and Weiss temperature, respectively), was applied to the $1/\chi$ vs T plot. The fit, however, failed to produce a convincible result.[16] Tentative CW parameters obtained in the calculations were considerably sensitive to least-squares fitting conditions, including temperature range width, and stable and reasonable solutions were never found. Further attempts were made to demonstrate the implied linear relationship between $1/\chi$ and T^2 for $SrRhO_3$. Neither the $T^{3/2}$ nor the $T^{4/3}$ fit (data not shown), however, yielded a linear part, which was expected, if $SrRhO_3$ was just at the magnetic instability point.[12,13] The above experimental observations would suggest that the magnetic susceptibility for $SrRhO_3$ is rather uncommon among properties of magnetic metals, because many antiferro- and ferromagnetic metals are expected to follow approximately the CW law above the magnetic ordering temperature or 0 K (in the case for nearly ordered metals).[11–13,17–20] The roughly estimated $\chi(0) \sim 1 \times 10^{-3}$ $\mathrm{cm^3/mol}$ for $SrRhO_3$ and the Sommerfeld constant discussed later ($\gamma = 7.6$ $\mathrm{mJ/mol\,K^2}$) yielded a Wilson ratio (R_{W}) of ~ 8.6 using the formula,[21]

$$R_{\mathrm{W}} = \frac{3\pi^2 k_{\mathrm{B}}^2 \chi(0)}{\mu_{\mathrm{B}}^2 \gamma}. \tag{1}$$

The preliminary R_{W} for $SrRhO_3$ is clearly out of the expected range, 1–2, for standard Fermi-liquid behavior. The unreliable R_{W} might support the presence of peculiar magnetism in $SrRhO_3$.

The most advanced profiling thus far achieved for the nearly and weakly antiferro- and ferromagnetic 3D metals was accomplished by developing the SCR theory of spin fluctuations in metals.[17] At the paramagnetic region, $1/\chi$ is expected to be in direct proportion to the dth power of T, where $d = 1$–$3/2$ and 1–$4/3$ for antiferro- and ferromagnetic 3D metals, respectively.[13] This is the most notable point to distinguish the progress of understanding in magnetism of metals achieved by the SCR studies, and so far observations, indeed, seem to be in the range ($1/\chi \sim T^d$).[18] The rather conventional Stoner's model ($1/\chi \sim T^2$) is far beyond the range. The rhodium oxide metal, however, shows a nearly T^2 dependence of $1/\chi$, which ironically matches the Stoner expectation.[17] Although the T^2 trend in $1/\chi$ was also predicted by a random phase approximation theory, using it here to analyze the present data may be problematic, because it is too limited in temperature range (only effective within extremely low temperature), due to mainly a lack of self-consistency.[17,22] Further considerations with additional studies may be necessary to conclusively determine the microscopic origin of the $1/\chi \sim T^2$ trend in $SrRhO_3$.

The specific heat data are presented in Fig. 5. A standard relaxation technique was employed in the measurement. The temperature dependence of the specific heat (C_{p}) of $SrRhO_3$

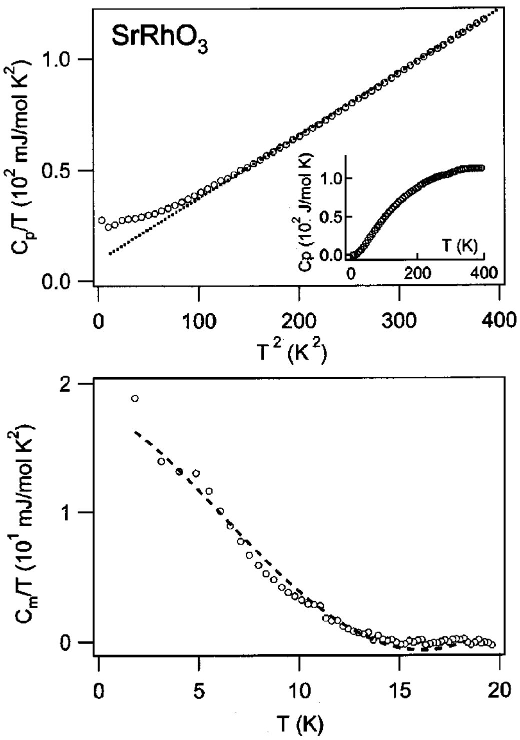

FIG. 5. Specific heat data for the polycrystalline $SrRhO_3$ (top) and the data after the orthorhombic lattice contribution and the Sommerfeld constant are subtracted (bottom). The size of error bars are as small as the circles. The estimated Debye temperature is 190 K and the Sommerfeld constant is 7.6 mJ/mol K^2 by a least-squares fitting as indicated by the dotted line. The dashed curve in the bottom panel is computed (see text).

measured between 1.8 and 390 K is plotted in the inset of the top panel in Fig. 5 after subtraction of a contribution from the addenda. The difference between C_p and C_v was assumed to be insignificant in the temperature range studied. The top main panel shows a C_p/T vs T^2 plot of the data below 20 K. As expected within the Debye approximation, a linear dependence is clearly seen. The estimated Debye temperature was 190 K and the Sommerfeld constant was 7.6 mJ/mol K^2 by a least-squares fitting as indicated by the dotted line in Fig. 5. Among Fermi-liquid metals, a universality was found in A/γ^2.[23] A tentative calculation of A/γ^2 with the obtained parameters for $SrRhO_3$, $\gamma = 7.6$ mJ/mol K^2 and $A = 0.062\mu\Omega$ cm/K^2, produced an incredible result, a value approximately two magnitudes larger than the universal constant. The parameter A for $SrRhO_3$ perhaps involves extrinsic contributions somewhat as ρ_0 does. We decided, therefore, not to make further quantitative analysis for A/γ^2 of $SrRhO_3$. On the other hand, we found that the Debye temperature of $SrRhO_3$ is much lower than those of the ruthenium oxide perovskites.[3] This fact would indicate that the lattice of $SrRhO_3$ is much "softer" than that of the ruthenium oxide perovskites. As expected from the Debye temperature, even within the studied temperature range, it can be clearly seen that the specific heat is approaching the roughly expected value ~125 mJ/mol K [5 (atoms per unit cell) ×3 (dimensionality per atom) $\times k_B N$ (Boltzmann's and Avogadro's constants)].

In the low-temperature portion of the specific heat data, an extra contribution (C_m) appears, as C_p/T starts to part gradually from linear dependence on cooling. It is presumably magnetic in origin and is found in a variety of itinerant magnetic materials.[3,24] The probable magnetic term was extracted by subtracting the lattice contribution and the Sommerfeld constant from the original data, as shown in the bottom panel of Fig. 5. At first, the C_m/T data were quantitatively investigated with a component for spin fluctuations in the SCR framework for nearly ferromagnetic metals.[3,20] The contribution of the spin fluctuations to the specific heat was approximated by

$$\frac{C_m}{T} \sim \frac{9N_0}{T_0}\int_0^{1/K_0} dx\, x^2 \frac{1}{t}\left[-u - \frac{1}{2} + u^2\Psi'(u)\right], \tag{2}$$

where N_0 is the number of magnetic atoms, $\Psi'(u)$ is the first derivative of the digamma function, T_0 and K_0 are the parameters as to spin fluctuations, $u = x[x^2 + \chi(0)/\chi]/t$, $t = T/T^*$, and $T^* = T_0/K_0^3$.[20] The expression was then reduced to the following form in the low-temperature limit:

$$\rightarrow \frac{3N_0}{4T_0}\left[\ln(1+K_0^{-2}) + \frac{2}{5}t^2 \ln t + \cdots\right]. \tag{3}$$

For fitting purposes by a least-squares method, T_0, K_0, and T^* were set as independent variable parameters in the first two terms in Eq. (3), where t was replaced by T/T^*. The best fitting result is shown in the bottom panel of Fig. 5 as a dashed curve. Although the observed data C_m/T vs T were reproduced at a convincible level, all of the parameters determined here, $T_0 = 0.003\,05$ K, $K_0 = 3.69$, and $T^* = 26.7$ K, were, however, incredible.[20] For example, the tentatively obtained values do not satisfy the form $T^* = T_0/K_0^3$ at all. As dictated by Eq. (3), there were no other combinations of the parameters that fit the data. These facts, therefore, suggest that the contribution from spin fluctuations in nearly ferromagnetic metals is either unlikely or at least insufficient to account for the observed C_m in $SrRhO_3$. In 3D nearly antiferromagnetic metals, magnetic contributions to the specific heat in the SCR framework have been studied; an enhancement of γ is expected at low temperature instead of the parameters in Eq. (3).[25] The 3D nearly antiferromagnetic picture is, therefore, unlikely to explain the observed C_m for $SrRhO_3$.

IV. CONCLUSIONS

The structure and electronic properties of a polycrystalline sample of $SrRhO_3$ obtained by high-pressure synthesis techniques were investigated. Although the polycrystalline nature of the sample limited quantitatively a detailed analysis of the electrical resistivity properties, the present data strongly suggest the perovskite is in the category of a Fermi liquid. The magnetic susceptibility of $SrRhO_3$ was found to follow a rather unusual temperature dependence, i.e., $1/\chi \sim T^2$. A tentative attempt at a quantitative analysis using 3D spin fluctuation models resulted in unconvincible results for the magnetic susceptibility and the specific heat data. Although the major contribution to the enhancement of the paramagnetism of $SrRhO_3$ might result from a sizable density of states at the Fermi level, as in $(Sr,Ca)RuO_3$,[2–4] it is not sufficient to explain the entire magnetic behavior of $SrRhO_3$, because it is temperature independent. There are likely additional factors which account for the temperature-dependent portion of the magnetism with the $1/\chi \sim T^2$ trend. The character of the paramagnetism of $SrRhO_3$ seems to be intermediate between that of enhanced Pauli- and CW-type paramagnetism. While extensive studies were made on paramagnon contributions for the CW paramagnetism in the vicinity of the critical point, intermediate paramagnetism was essentially uninvestigated. Whether the rhodium oxide 3D metal tends toward either an antiferro- or ferromagnetic instability point, the imposing appearance of the distinctive T^2 term in $1/\chi$ indicates that the magnetic excitation of 4*d* electrons in $SrRhO_3$ remains highly elusive. Further investigations into $SrRhO_3$, including theoretical considerations, would be of significant interest.

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