Large magnetoresistance of the ferromagnetic intermetallics URhSi and URhGe

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URhSi and URhGe have been studied by means of magnetization and magnetoresistance measurements. These compounds undergo a magnetic phase transition into the ferromagnetic state at 9.5 and 9.0 K, respectively. A large magnetoresistance near their Curie temperature is observed. At $B = 8$ T, the maximum magnetoresistance is -36% at 10.3 K for URhSi and -40% at 9.5 K for URhGe. We propose that this large magnetoresistance is a consequence of two kinds of scattering: the spin-disorder scattering and scattering from magnetic polarons. [S0163-1829(98)03718-7]

I. INTRODUCTION

The equiatomic ternary uranium compounds UTM ($T = d$-electron transition metal, $M =$ Si or Ge) crystallize in either the CeCu$_2$-type structure (space group $Imma$) or its ordered version of the TiNiSi type (space group $Pnma$).[1–3] In these compounds a hybridization between the $5f$ electrons and d electrons derived from the transition metal T plays a very important role in their magnetic properties. Hence, the members of this series exhibit a variety of properties depending on an involved T atom. It was observed that the compounds with transition metals having more filled d states (UNiM, UPdM, UPtM) order antiferromagnetically.[2–6] On the other hand, the compounds with a smaller number of d electrons (UFeM, UCoM, URuM, UIrSi) are commonly spin fluctuators or paramagnets to the lowest temperatures measured.[2–6] Thus, the special place in the UTM series is occupied by URhSi and URhGe for which we have already reported the magnetic and electrical resistivity measurements.[2,4] From the magnetization measurements it follows that these two materials undergo a ferromagnetic phase transition with $T_C = 9.5$ and 9.0 K, respectively. The electrical resistivity ρ of URhSi and URhGe shows a smooth dependence with temperature above T_C and a knee at T_C. A strong drop in the resistivity below T_C is well known due to the ferromagnetic ordering in the samples. Specific heat measurements on URhSi and URhGe were performed by de Boer *et al.*[5] and Buschow *et al.*,[6] respectively. The temperature indicating a ferromagnetic phase transition determined from this experiment in both cases showed good agreement with the results of magnetic and electrical measurements.

The most salient property in these URhM alloys is that they exhibit features corresponding rather to an itinerant $5f$-electron magnetism, i.e., the low value of magnetic moment and the lack of saturation in magnetization.[2] This behavior is supported by the high-field magnetization measurements where no full saturation is even achieved up to 40 T.[7] Further support in this regard has been obtained from measurements of the specific heat mentioned above.[5,6] Its linear term γ is characterized by a large value of about 150 mJ/mol K^2, which indicates the importance of the band effects in these materials.

Up to now, however, little attention has been given to the crystal structure, transport properties, and magnetic structure of the URhM compounds. Therefore, we have undertaken a neutron powder diffraction study,[8] and performed more detailed magnetization and magnetoresistance measurements on URhSi and URhGe at low temperatures. The neutron diffraction results have indicated that these intermetallic compounds crystallize in the orthorhombic structure of the TiNiSi type with the space group $Pnma$. Below T_C both these compounds undergo a magnetic phase transition into the itinerant ferromagnetic state with the same magnetic space group ($Pn'm'a$). The present work focuses mainly on the transport properties of the URhSi and URhGe compounds and has revealed properties unknown so far that these materials exhibit a huge value of magnetoresistance near their Curie temperatures.

II. EXPERIMENTAL DETAILS

The materials were prepared by arc melting stoichiometric constituents under a high-purity argon atmosphere. Weight losses were checked to be within 1% mass. After an annealing at 800 °C for two weeks, the x-ray analysis revealed that the samples were single phase with an orthorhombic structure. The prepared samples by us were subjected to magnetization and magnetoresistance measurements.

The magnetization measurements were performed on the bulk sample between 1.7 and 25 K in magnetic fields up to 5.5 T, using a superconducting quantum interference device magnetometer (Quantum Design MPMS-5 type). Between each field sweep the sample was warmed to the temperature just slightly above 100 K and then it was zero-field cooled to the next measuring temperature.

Electrical resistivity (ρ) measurements were performed using standard four probe dc technique. The temperature dependence of ρ(T) from 1.5 to 300 K was measured in zero field and between 4.2 and 100 K in a fixed magnetic field of 8 T. In turn, the field dependence of the resistivity $\rho(B)$ was measured at several temperatures in the range 4.2 – 25 K in increasing fields up to 8 T and followed decreasing fields. The fields always were applied along the longest sample dimension. The longitudinal magnetoresistance (MR) then is defined as $\Delta\rho/\rho = \{\rho(B,T) - \rho(0,T)\}/\rho(0,T)$. In these experiments a current of 10 mA was used and uncertainty in
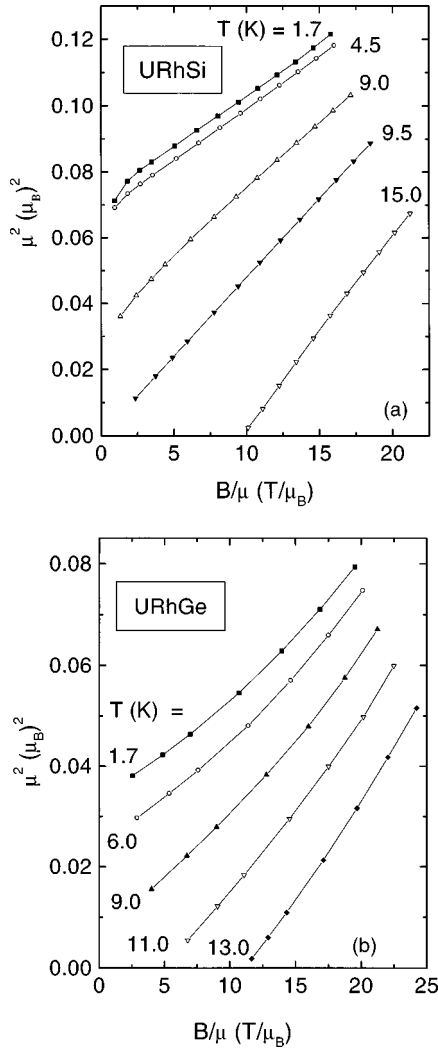
FIG. 1. Magnetization data plotted as μ^2 versus B/μ for (a) URhSi and (b) URhGe.

the geometrical factor was less than 5%. The voltage measurements were made automatically by a personal computer connected with a Keithley 181 nanovoltameter. The temperatures below 4.2 K were achieved by pumping over the liquid-helium. The temperature of the sample was controlled and measured by a Lake Shore carbon glass sensor, employing a temperature controller (Oxford ITC 503 type).

III. RESULTS AND ANALYSIS

For URhSi and URhGe several isothermal magnetization curves were taken at temperatures below 15 K. The magnetization at 1.7 K shows only a slight tendency to saturate in a maximum applied field of 5.5 T. When the temperature is raised, the magnetic response at 5.5 T slowly decreases. The spontaneous magnetization obtained from an Arrott plot (μ^2 versus B/μ) (Fig. 1) is very small, i.e., 0.27 for URhSi and 0.19μ_B for URhGe at 1.7 K. These values are two times smaller than the ordered moments determined by our neutron diffraction study,[8] and about three times smaller than the values of magnetic moments determined by high magnetic field measurements up to 35 T carried out by de Boer et al.[7] on the oriented in magnetic field powder samples. Our mag-
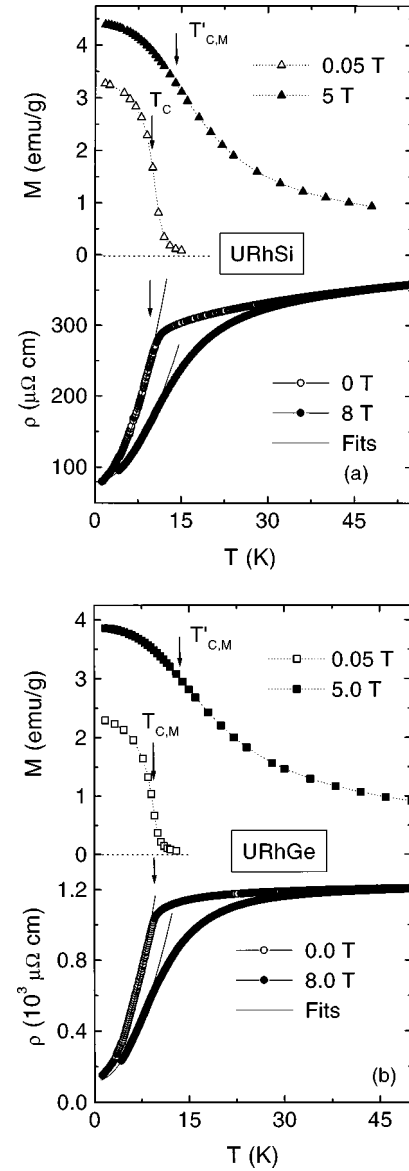

FIG. 2. Temperature dependence of the magnetization $M(T)$ and of the electrical resistivity, measured in various applied magnetic fields up to 8.0 T for (a) URhSi and (b) URhGe. The arrows indicate the Curie temperature. The solid lines are fits to the experimental data.

netization values are rather close to those measured by these authors on the sample with randomly oriented grains frozen in alcohol.

The Curie temperature T_C deduced from the Arrott plot amounts to 9.5 and 9.0 K for URhSi and URhGe, respectively. For URhSi, the Arrott plot reveals μ^2 versus B/μ straight lines. On this basis we have inferred a value of T_C for this compound. The dependences for URhGe are rather curvature and the procedure of an extrapolation in this case is less reasonable. Nevertheless, the determined values T_C are in agreement with our previous studies and correspond to the minimum in the temperature derivative of the magnetization measured in very low magnetic fields, denoted here as $T_{C,M}$ (an inflection point). Upon application of a magnetic field of 5 T, the temperature of the inflection point $T'_{C,M}$ grows almost double (Fig. 2). This behavior may indi-

TABLE I. Electron-transport and magnetic properties of URhSi and URhGe. T_C is the Curie temperature deduced from the Arrott plots. $T_{C,M}$ and $T'_{C,M}$ are the temperatures at which the derivative of the magnetization $dM(T)/dT$, measured at 0.05 and 5.0 T shows a minimum. $T_{C,\rho}$ is the temperatures corresponding to maximum of the derivative $d\rho(T)/dT$. T_m is the temperature corresponding to the $\Delta\rho/\rho$ maximum at $B = 8$ T. $\Delta\rho/\rho$ is the maximum value of magnetoresistance at T_m.

Compounds	T_C (K)	$T_{C,M}$ (K)	$T'_{C,M}$ (K)	$T_{C,\rho}$ (K)	T_m (K)	$\Delta\rho/\rho$ (%)
URhSi	9.5	10.0	14.0	9.5	10.3	−35
URhGe	9.0	9.2	13.5	9.0	9.0	−40

cate the existence of magnetic interactions between uranium atoms even far above the ferromagnetic phase transition. The Curie temperatures determined by magnetization measurements together with $T'_{C,M}$ are listed in Table I.

In Fig. 2 we report the electrical resistivity data of URhSi and URhGe as a function of temperature measured in zero field $[\rho(0,T)]$ and in 8 T $[\rho(8,T)]$. In the paramagnetic state, the resistivity of URhSi slightly decreases with lowering temperature, while for URhGe $\rho(T)$ is practically independent of T. This behavior suggests the existence of an additional contribution to the resistivity, since the two well-known contributions, i.e., a constant contribution of the spin-disorder scattering and an increasing with temperature contribution of the phonons, should not lead to a leveling off of the temperature dependence of the resistivity above T_C. Therefore, there is an additional contribution of electron scattering one should probably take into account, such as, for example, magnetic polaron-type, which will be discussed below.

At low temperatures, the zero-field $\rho(0,T)$ function of both investigated compounds is characteristic of a ferromagnetic material, for which the change with decreasing temperature below T_C is governed by a gradual decrease of the spin-order scattering. The temperature of the anomaly observed in the $\rho(0,T)$ curves is well reflected by a maximum in the derivative of resistivity $d\rho(T)/dT$ and corresponds well to the respective T_C point determined from the magnetic measurements (see Table I). The low-temperature part of the resistivity can be analyzed by the equation

$$\rho(T)=\rho_0+AT^2, \qquad (1)$$

where ρ_0 is the temperature-independent residual resistivity and AT^2 is the resistivity due to scattering by magnons. A least-squares fit of the data taken between 1.5 and 7 K to Eq. (1) gives $\rho_0 = 80.3\ (\pm0.3)$ and $132\ (\pm1)$ $\mu\Omega$ cm and $A = 1.84\ (\pm0.01)$ and $11.1\ (\pm0.1)$ $\mu\Omega$ cm/K^2 for URhSi and URhGe, respectively. The quadratic temperature dependence of the resistivity measured at 8 T $\rho(8,T)$ is also evident below 7 K. The fits of experimental data to Eq. (1) are shown in Fig. 1. One can see that the values of the coefficient A are smaller [accounting to 0.9 (±0.1) and 6.4 (±0.1) $\mu\Omega$ cm/K^2 for URhSi and URhGe, respectively). However, due to the rather narrow range of temperatures measured the absolute values inferred from the fits should be taken with large care. As expected for ferromagnetic materials, the derived ρ_0 values from these fits of the experimental data are close to those found from the $\rho(0,T)$ data. Thus, in the low-temperature regime, the scattering by magnons appears to be responsible for the resistivity behavior. With increasing tem-

perature, the effect of applied magnetic field of 8 T on the resistivity becomes significant. For both URhSi and URhGe, the accompanying resistivity change gives rise to an enormous magnetoresistance change.

The most remarkable point is the magnitude of magnetoresistance which reaches its maximum at the temperature T_m (Fig. 3), where the derivative $d\rho(8,T)/dT$ shows also a maximum. At T_m, $\Delta\rho/\rho$ is as large as −36% for URhSi and −40% for URhGe. This huge change in $\Delta\rho/\rho$ of these materials is distinctly seen in the data collected in a wide temperature range, even at temperatures well higher than T_C.

In Fig. 4 we show the magnetization and magnetoresistance of URhSi and URhGe at several temperatures below 25 K and in fields up to 5.5 and 8 T, respectively. The measurements were made in increasing and decreasing fields. Thus, it is clear from this figure that $\Delta\rho/\rho$, as with magnetization, shows no hysteresis effect. Below T_C, both the magnetization and magnetoresistance do not reach saturation in the maximum fields applied here. In the paramagnetic state, the $M(B)$ plots for these two compounds display a small curvature giving rise to the nonlinearity of this dependence in the high field region. At the same time $\Delta\rho/\rho$ taken at 25 K, i.e., at a temperature much higher than the relevant T_C of these studied compounds, depends on B nearly quadratically and is still negative. Although such a negative contribution is often
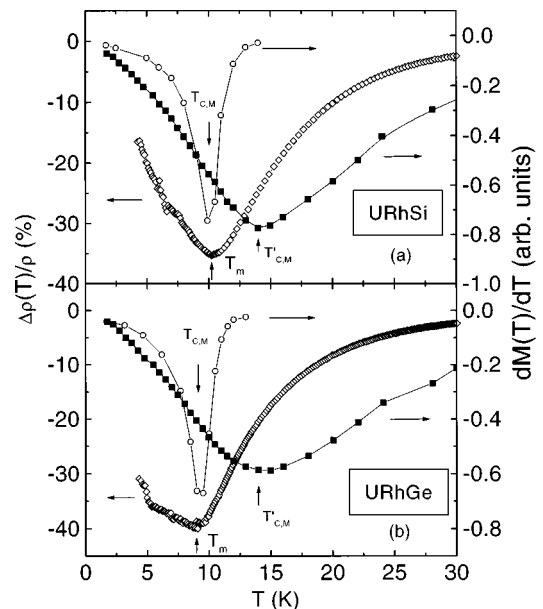

FIG. 3. Temperature dependence of the magnetoresistance $\Delta\rho/\rho$ at $B=8$ T (open diamonds) and derivatives of the magnetization dM/dT, measured at 0.05 (open circles) and 5 T (solid squares) for (a) URhSi and (b) URhGe.
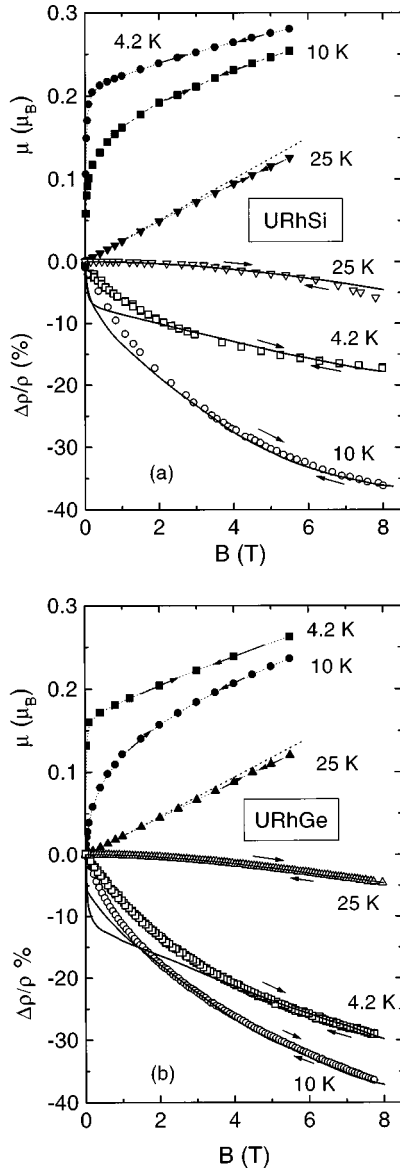
FIG. 4. The effect of a magnetic field on the magnetization and magnetoresistance measured at several temperatures below 25 K for (a) URhSi and (b) URhGe. The solid lines are fits to the experimental data.

characteristic of a Kondo effect, here this feature probably comes from the polarization of the local uranium moments. These experimental findings together with the data presented in Fig. 2 lead us to a suggestion that the magnetoresistance and the magnetization are closely related to each other. Therefore, we have tried to establish this relation using the effective exchange interaction model (EEI) given by Van Peski-Tinbergen and Dekker.[9] This model has been successfully applied in an analysis of the giant magnetoresistance (GMR) of the Fe-based binary systems Fe_xM_{1-x} ($M=$Cu, Ag, Au).[10] In this series relatively large MR values of up to 15–30 % were obtained for each M with $x=0.2$, 0.13, and 0.15, respectively. As with our case, the MR of these alloys does not reach saturation even at the highest measured field of 8 T.

According to the EEI model the measured resistivity $\rho(B,T)$ is expressed as

$$\rho(B,T)=\rho(0,T)\{[1+a\ \tanh\ (g\mu_B B/2k_B T)M(B,T)]$$
$$-c^2 M^2(B,T)/[1+a\ \tanh$$
$$\times(g\mu_B B/2k_B T)M(B,T)]\},\qquad(2)$$

where a and c are both constants, all connected with the spin-independent scattering potential V, the effective exchange interaction J, and the spin $\bar{S}$ of a magnetic scatterer. We have applied this equation to fit the observed data with a and c as adjustable parameters. The results of fitting as solid lines are shown in Fig. 4. As one can expect, the observed deviations at low fields and low temperatures result in the fact that the EEI model is more applied for isotropic systems. In general, due to the $5f$ electrons the magnetization of almost all known intermetallic uranium compounds is more or less anisotropic. Hence, the derived here theoretical curves can fairly well describe only part of the experimental data, especially taken at high fields and at temperatures more closed to T_C. Nevertheless, on the basis of the obtained results, it is quite indicative that the magnetoresistance may be a unique function of magnetization, confirming a strong coupling of the charge carries with the uranium magnetic moments.

IV. DISCUSSION

From a classical point of view, the behavior of URhSi and URhGe, showing a negative minimum in $\Delta\rho/\rho$ near T_C, is expected for normal metallic ferromagnets, for which the spin-disorder scattering can be suppressed by an applied magnetic field.[11,12] However, there are two important facts which clearly do not offer strong evidence for this type of scattering in our compounds. First, the spin-disorder scattering in metallic ferromagnets is usually weak to give a marked effect in $\Delta\rho/\rho$ (2% for a typical ferromagnet Ni). In our compounds, the observed magnetoresistance is rather significant ($\Delta\rho/\rho > -35\%$), and therefore, at least of an order of magnitude higher than what one expects in any metallic ferromagnet. Second, we have found that the change in $\Delta\rho/\rho$ does not disappear even in a wide temperature region above T_C. The latter behavior cannot be explained in terms of spin-disorder scattering, since the resistivity of such a scattering on the disordered magnetic moments just above the magnetic phase transition becomes independent of temperature. On the basis of these experimental findings, we think that apart from the minor contribution originating from the spin-disorder scattering, an additional strong contribution to $\Delta\rho/\rho$ starts to show up. This contribution is negative and becomes a determining factor thus in the temperature range above T_C.

To explain the magnetoresistance behavior of the URhM ternaries one should take into account the following features: (i) the observed $\Delta\rho/\rho$ behavior is related to the $5f$-electron interactions, (ii) the mechanisms leading to a significant $\Delta\rho/\rho$ value at T_C itself, and (iii) a large change in $\Delta\rho/\rho$ occurs at the temperature close to T_C and just above it. Unfortunately, a coherent theory which could explain these observations is not available at present.

Let us consider some groups of compounds which show a GMR effect. One such group we should mention is that of the antiferromagnetic $5f$-electron compounds such as

UNiAl,[13] UNiGa,[14,15] UPdIn,[16] UNiGe,[17] and UPdGe.[18] In these intermetallics the large values of $\Delta\rho/\rho$ arise mainly from the marked change in $\rho(B)$ due to the field induced transition from the antiferromagnetic to ferromagnetic state. In addition, both the magnetization and magnetoresistance of the materials mentioned above, except for UNiAl, saturate rapidly above the critical field B_{cr}. On the contrary, here, we deal with ferromagnetic compounds and despite the exhibition of the large magnetoresistance at 8 T they do not approach a saturation in this field; they are far from it. This gives one an argument that the explanation of this behavior in terms of previously proposed mechanisms,[19] i.e., the drastic changes in the Fermi surface topology as an origin of the GMR effect for the antiferromagnetic materials mentioned above, cannot be applied in our case.

In other classes of materials, namely, in magnetic multilayers,[20] and in granular heterogeneous Cu-Co and Ag-Co magnetic alloy films,[21] the GMR effect has previously been observed as well. For these layers, the occurrence of the GMR effect is usually ascribed to the spin-dependent scattering.[20] This appears because of the resistivity being dependent on the relative magnetic orientation of neighboring ferromagnetic layers. The resistivity is higher for an antiparallel alignment of magnetic layers compared to a parallel alignment. Moreover, there are theoretical calculations which predict a strong influence of the electronic structure on the GMR (for references, see Ref. 22). In any case, the observed GMR effect relates to a layered structure, which is determined by an interface roughness and some randomness in the lattices. There is an essential difference compared with the compounds investigated here, where the uranium atoms are located on the periodic lattice sites. Hence, the spin-dependent scattering does not occur in our materials.

Recently, the Mn-based oxides $L_{1-x}A_x$MnO$_3$ (L=La, Nd, Pr, etc., and $A=$ Ca, Sr, Ba, etc.) have been studied intensively due to the fact that they show a colossal magnetoresistance of an order of 10^5%.[23] A common crystal structure to all of these manganites is the perovskite one. These compounds are isolators in the paramagnetic state but become metallic in the ferromagnetic state. For $T>T_C$, the negative GMR has been explained as being due to a hopping of magnetic polarons.[24] In this feature, the magnetic polarons are formed near the ferromagnetic phase transition and they are associated with some orientation of the spin clusters of the pairs of Mn^{3+} and Mn^{4+} ions.

Bearing in mind that the mechanism causing the GMR may not be the same in various systems, but among the mechanisms proposed here for our URhM materials, the contribution of the magnetic polarons[24] as an origin of the GMR effect seems to be possible as well.

Because of the existence of short-range order far above T_C, we assume that the magnetic correlations between the uranium moments form some oriented spin clusters at temperatures more or less close to T_C. The presence of magnetically inhomogeneous regions at these temperatures owing to such spin clusters must bring about the different type of electron scattering in the sample. One of them will be coherent if the magnetic scattering is just associated with the magnetic polarons. This scattering should be effective when the electron moves in a region of magnetic coherence length smaller than the electronic free mean length. In our sample, this type of scattering can happen at extended temperatures as high as $5T_C$ and thus, the weak temperature dependence of the resistivity in this temperature range is consistent with this feature. At T_C and at lower temperatures, the polarons are no longer formed because the uranium magnetic moments become long-range ordered. Also the applied magnetic field reduces the number of polarons owing to the increasing degree in the magnetic order. Therefore, the resistivity starts to drop when the polaron scattering begins to be reduced strongly by the external magnetic field. Here, due to the additional spin-disorder scattering there is a rapid falloff in the resistivity below T_C. Finally, if this proposed mechanism is true for our URhM compounds, the large magnetoresistance effect can originate from the short-range magnetic order on the scale of the mean free path the charge carries. However, we should note that this is one of the proposed models for explaining the transport properties of the ferromagnetic intermetallics URhM, but further studies need to be done to confirm it. Very recently, such a view on GMR has been proposed by Mallik *et al.*[25] for an intermetallic ferromagnet GdNi.

V. CONCLUSION

Our measurements have indicated that the magnetoresistance of the itinerant ferromagnets URhSi and URhGe at 8 T and at T_C reaches a large value and decays in a wide temperature range even up to 50 K (five times above T_C). As was proposed in Ref. 25, we also attribute the gradual increase of negative magnetoresistance below the temperature as high as $5T_C$ to magnetic scattering arising from some kind of magnetic polaron. Apart from this mechanism, we have also discussed several other mechanisms leading to a large MR effect. Nevertheless, the proper mechanisms responsible for the magnetoresistance effect observed in URhM have not yet been definitely identified.

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