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Crossing point phenomena (T* = 2.7 K) in specific heat curves of superconducting ferromagnets $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10\text{-}\delta}$

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Crossing point phenomena are one of the interesting and still puzzling effects in strongly correlated electron systems. We have synthesized $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10\text{-}\delta}$ (GdRu-1222) magneto-superconductor through the standard solid state reaction route and measured its magnetic, transport, and thermal properties. We also synthesized $RuSr_2Eu_{1.4}Ce_{0.6}Cu_2O_{10\text{-}\delta}$ (EuRu-1222) then measured its heat capacity in zero magnetic fields for reference. The studied compounds crystallized in the tetragonal structure with space group *I4*/*mmm*. To explore the crossing point phenomena, the specific heat [C_p (T)] was investigated in the temperature range 1.9-250 K, under magnetic field of up to 70 kOe. Unfortunately though no magnetic and superconducting transitions are observed in specific heat, a Schottky-type anomaly is observed at low temperatures below 20 K. This low temperature Schottky-type anomaly can be attributed to splitting of the ground state spectroscopic term $^8S_{7/2}$ of paramagnetic Gd^{3+} ions by both internal and external magnetic fields. It was also observed that C_p (T) is measured for different values of magnetic field, possesses the same crossing point ($T^* = 2.7$ K), up to the applied magnetic field 70 kOe. A quantitative explanation of this phenomenon, based on its shape and temperature dependence of the associated generalized heat capacity (C_p), is presented. © *2012 American Institute of Physics*. [doi:10.1063/1.3677868]

I. INTRODUCTION

Discovery of the co-existence of superconductivity and magnetic order in the hybrid rutheno-cuperate systems $RuSr_2(Eu,Gd,Sm)_{1.5}Ce_{0.5}Cu_2O_{10\text{-}\delta}$ (RERu-1222) and $RuSr_2(Eu,Gd,Sm)Cu_2O_{8\text{-}\delta}$ (RERu-1212) is interesting because the magnetic ordering temperature is much higher than the superconducting transition temperature.[1–5] However, despite the extensive research on these materials, some questions yet remain unanswered. The first simultaneous observation of superconductivity and the Ru ion magnetic order in a rutheno-cuprate was published in 1997 by Felner *et al.*[1] These systems show ferromagnetic ordering at a higher Curie temperature of around 100-135 K and superconductivity at a lower critical temperature of about 15-40 K. The superconductivity is associated with the $Cu\text{-}O_2$ planes, while the magnetic order originates from the $Ru\text{-}O_2$ planes. These $Ru\text{-}O_2$ planes are not only responsible for weak ferromagnetism but also provide charge carriers to the superconducting $Cu\text{-}O_2$ planes.

Very recently, a crossing point phenomenon is seen due to ground state spectroscopic term splitting of Gd^{3+} ion in heat capacity of GdRu-1222.[6] The crossing point phenomenon is one of the interesting and still puzzling effects in strongly correlated electron systems.[7–9] A typical example of this effect is the temperature dependence of specific heat curves C_p (T, X) observed at different values of a thermodynamic variable X (such as magnetic field H or pressure P). This type of effect was found not only for thermodynamic but for dynamical quantities, i.e., frequency dependent optical conductivity, etc. Generally, this familiar effect is named as the *isobestic point*.[10] The curves cross at one particular temperature known as crossing point temperature T*. In particular, the crossing point in C_p (T, H) curves was found in the heavy-fermion compounds like $CeCu_{5.5}Au_{0.5}$,[11] semi-metallic $Eu_{0.5}Sr_{0.5}As_3$,[12] superconducting cuprate $GdBa_2Cu_3O_{7\text{-}\delta}$,[13] and manganite $NdMnO_3$.[14] The known theoretical considerations[7–10] are based on rather different approaches. Available relevant experimental data are inconclusive, therefore more experimental data in support this phenomenon are warranted for different strongly correlated electron systems. Until now the general reasons and conditions for occurrence of *isobestic points* are not clear. In this report, the crossing point effect is shown in C_p (T, H) curves of polycrystalline known superconducting ferromagnets $RuSr_2\ Gd_{1.4}Ce_{0.6}\text{-}Cu_2O_{10\text{-}\delta}$ (GdRu-1222). An interesting phenomenon associated with the crossing point is the Schottky anomaly, where the specific heat capacity of a solid at low temperature makes a "*bump*." It is called anomalous because the heat capacity usually increases with temperature or remains constant.[15]

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II. EXPERIMENTAL DETAILS

Polycrystalline bulk samples of $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ (GdRu-1222) and $RuSr_2Eu_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ (EuRu-1222) are synthesized through standard solid state reaction route. More experimental detailed are given in Ref. 1 and 3. X-ray diffraction (XRD) of studied GdRu-1222 was done at room temperature in the scattering angular (2θ) range of 20°-80° in equal steps of 0.02° using the Rigaku diffractometer with CuK_α ($\lambda = 1.54$ Å) radiation. Detailed Rietveld analysis was performed using the FullProf program. DC magnetization, DC transport for GdRu-1222 and specific heat measurements over the temperature range 1.9-250 K for GdRu-1222 and EuRu-1222 were performed on PPMS, 14T Quantum Design USA.

III. RESULTS AND DISCUSSION

As seen from Fig. 1, characteristic impurity peaks corresponding to $SrRuO_3$ and Sr_2GdRuO_6 phases are not observed and rather a phase pure GdRu-1222 is seen within the resolution limit of XRD. Observed and fitted X-ray pattern for the compound is shown in Fig. 1. The structural analysis was performed using the Rietveld refinement analysis by using the FullProf Program. All the Reitveld refined structural parameters (lattice parameters, site occupancy, and atomic coordinates) of the compound are shown in Table I.

TABLE I. Atomic coordinates and site occupancy for studied $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$
Space group: ***I4/mmm***, Lattice parameters; ***a* = 3.8352 (4) Å, *c* = 28.5728 (3) Å, χ^2 = 3.76**.

Atom	Site	x (Å)	y (Å)	z (Å)
Ru	*2b*	0.0000	0.0000	0.0000
Sr	*2h*	0.0000	0.0000	0.4205 (4)
Gd/Ce	*1c*	0.0000	0.0000	0.2942 (6)
Cu	*4e*	0.0000	0.0000	0.1455 (3)
O(1)	*8j*	0.6152 (3)	0.5000	0.0000
O(2)	*4e*	0.0000	0.0000	0.0741 (2)
O(3)	*8g*	0.0000	0.5000	0.1461 (3)
O(4)	*4d*	0.0000	0.5000	0.2500

The temperature behavior of magnetization, *M-T* shows important features of the complicated magnetic state of this compound. The main panel of Fig. 2 depicts the temperature dependence of zero-field-cooled (ZFC) and field-cooled (FC) DC magnetization plots for the studied $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ (GdRu-1222) magneto-superconductor, being measured at 20 Oe applied field. The sharp rise of both ZFC and FC curve at (Curie temperature) $T_C = 120$ K shows a paramagnetic (PM) to ferromagnetic (FM) transition. The system enters into a new phase called glassy phase[16] just below the Curie temperature (T_C). The ZFC curve shows a peak at around $T_f = 80$ K, just below the temperature, where the ZFC and FC curve separates. In the ZFC curve the system undergoes a superconducting transition at $T_c = 27$ K. The inset of Fig. 2 shows the temperature dependence of resistance of studied GdRu-1222 from the temperature range 10-300 K. As we move from high temperature to low temperature the resistance increases slightly exhibiting the insulator-like behavior. But just below the freezing temperature ($T_f = 80$ K), the resistance increases very fast and finally drops sharply at the SC transition at $T_c = 27$ K.

Figure 3 reveals specific heat at low temperature range (below 40 K) of studied GdRu-1222 and EuRu-1222 in zero magnetic fields. Interestingly no magnetic (~150 K) or superconducting transitions (~30 K) are observed in both the samples, see inset Fig. 3. Rather, an upturn in heat capacity C_p (T) below 20 K (Schottky-type anomaly) is observed for the GdRu-1222 sample. This low-temperature Schottky-type anomaly below 20 K can be attributed due to splitting of the ground state term $^8S_{7/2}$ of paramagnetic ions by internal and external magnetic fields. According to the Kramer's theorem, the degenerate ground state term should split into four doublets in tetragonal symmetry. Internal molecular fields can arise in rutheno-cuprate from both the Gd and Ru sublattice[17] and can coexist with superconductivity. Even though a direct Gd-Gd exchange interaction is unlikely, these ions can be magnetically polarized by the *4d-4f* interaction, as discussed in more details in Ref. 18. Generally, the Schottky term in the specific heat for the compounds with Gd^{3+} ions should be attributed to the splitting of all four doublets, although actually only some of them make the dominant contribution to the effect. The ground state term for Eu ions is[7] F_0, hence no splitting takes place on the

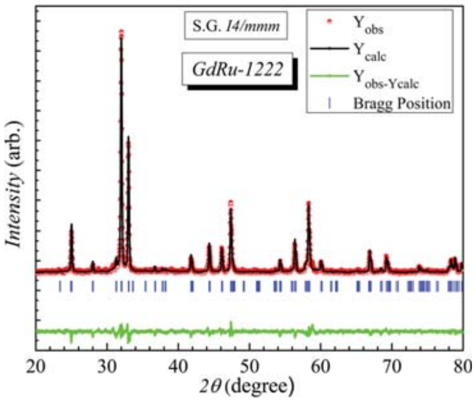

FIG. 1. (Color online) Observed (solid circles) and calculated (solid lines) XRD patterns of $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ compound at room temperature.

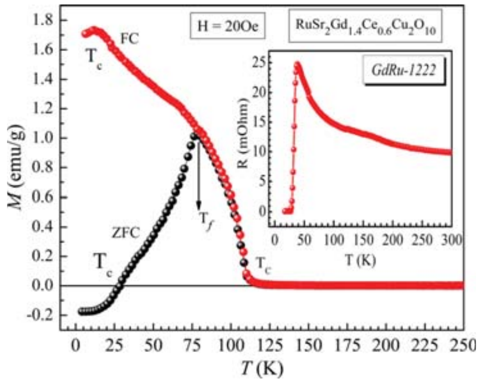

FIG. 2. (Color online) ZFC and FC DC magnetization plot for $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ measured in the applied magnetic field, H = 20 Oe. Inset shows the R vs T plot for O_2-annealed GdRu-1222.

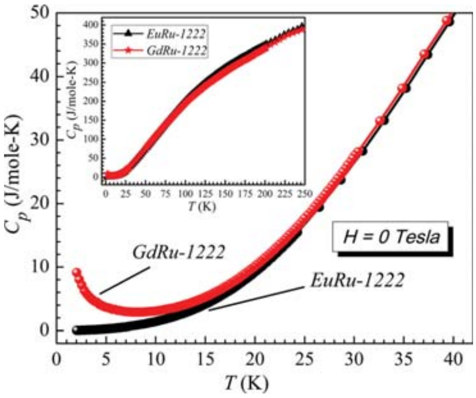

FIG. 3. (Color online) Low temperature behavior of total specific heat at zero magnetic fields for samples GdRu-1222 ($RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$) and EuRu-1222 ($RuSr_2Eu_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$). Inset shows heat capacity of the same for temperature range 1.9-250 K.

application of magnetic field. The EuRu-1222 sample shows smooth C_p (T) dependencies, the curve is of the Debye-type without any low temperature magnetic anomaly. Also, it should be noted that no magnetic transition (Curie temperature $T_C = 100$-120 K) was observed in heat capacity for both GdRu-1222 and EuRu-1222. This can be explained due to the absence of long range magnetic order in these systems.[19] There may be magnetic inhomogeneities as well at the transition point.

Figure 4 shows the specific heat curves for the GdRu-1222 sample observed at different values of applied magnetic field. All heat capacity curves cross at the same temperature point (known as crossing temperature) $T^* = 2.7$ K with the specific heat value $C_p{}^* = 7.04$ J/mol K. This happens only for the GdRu-1222 and not for EuRu-1222 sample. The crossing point effect is considered as some special type of universality for strongly correlated electron systems, but still no defined theory or mechanism for this phenomenon. Only some general reasons and preliminary thoughts are provided for it occurrence. It is mentioned in Ref. 10 that the crossing point appears in a system with superposition of two (or more) components, like that in the known Gorter-Casimir two fluid model of superconductivity. In consideration of the crossing effect in C_p (T, H) curves, it is important to know the motive force for strong magnetic field dependence of specific heat. For GdRu-1222, the motive force is associated with splitting of the ground state term $^8S_{7/2}$ of paramagnetic Gd^{3+} ions by internal and external magnetic fields.[18]

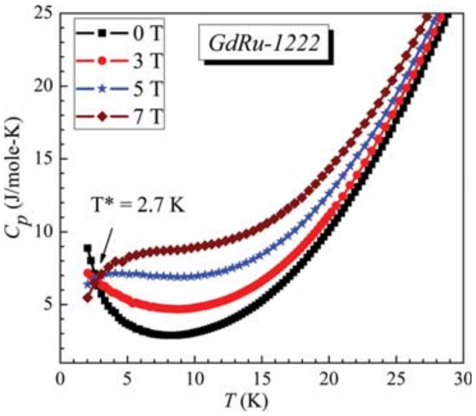

FIG. 4. (Color online) Low temperature dependencies of total specific heat (C_p), taken at different values of applied magnetic field for $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$.

The Schottky anomaly in the C_p (T, H) curves is itself a background for the crossing effect found in this study. There is a thermodynamics approach,[6] which can be helpful to understand this phenomenon. It can be suggested that in the low temperature range, where the crossing point phenomena takes place, the magnetic contribution to the specific heat is dominant. The expression for the specific heat at constant H is $C_H = T\,(dS/dT)_H$. Only if T^* is independent of H, will all C_H (T, H) curves intersect in one point demonstrating a true crossing effect (see Fig. 4).

In conclusions, both GdRu-1222 and EuRu-1222 are synthesized and measured for their structural, magnetic and heat capacity. The crossing point phenomenon in GdRu-1222 is observed and discussed. To compare the paramagnetic effects of different rare-earth compounds, we also measured magnetization and low temperature heat capacity of EuRu-1222. The low temperature Schottky-type anomaly is observed only in the GdRu-1222 compound and not in EuRu-1222. This is due to the splitting of the ground state term $^8S_{7/2}$ of paramagnetic ions Gd^{3+} by applied internal and external magnetic fields.

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