Interplay between disorder-driven non-Fermi-liquid behavior and magnetism in the Ce$_{0.24}$La$_{0.76}$Ge compound

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Introduction. – In the area of research on 4f electron-based compounds, a challenge which remains elusive is to understand the physical properties of materials where a singular interaction is mediated by soft collective modes. This investigation is important as the obtained results violate the applicability of the Fermi-liquid (FL) theory [1,2]. The FL theory forms a basis to understand the electronic properties and according to this theory, electrical resistivity ($\rho$) varies as the square of temperature ($T$) and the ratio of heat capacity ($C$) and temperature, i.e., $C/T$ is constant [3,4]. However, it has been noted in many 4f electron-based compounds that the FL theory breaks down and shows a non-Fermi-liquid (NFL) behavior near a quantum critical point (QCP). The $\rho$ of such compound shows a nonlinear behavior with temperature varying as $T^{1.5}$, while the $T$ variation of $C/T$ is logarithmic [5–8]. The observation of such NFL behavior might arise due to the presence of soft order parameter fluctuations resulting in singular interactions mediated between electrons [9–11]. In some compounds exhibiting NFL behavior, $\rho$ is reported to vary as $T^{0.5}$ [12–14]. The observation of such features is explained on the basis of the multi-channel Kondo model (such as the two-channel Kondo model), which is expected in 4f electron-based compounds [15–18]. The conduction electron is further coupled to the 4f electron state via the spin-orbit coupling which results in stretching and squashing of the conduction electron orbital. This effect might be responsible for the splitting of sixfold degeneracy of $J = 5/2$ ground multiplets into the quartet excited state along with the doublet ground state under crystalline electric field [17–21]. The coupling of the quartet of conduction electron with localized moments can be described by the two-channel Kondo model which results in the fact that $\rho$ varies with $T^{0.5}$ [22]. However, in a disorder-driven NFL compound, $\rho$ varies with $T^{1.6}$ [23–25]. For understanding this behavior of resistivity, recently, a spin glass (SG) model in the frame of the dynamical mean-field theory has been predicted [24,26]. This theory explains such variation in resistivity on the basis of the presence of disorder when the exchange energy is comparable to the Kondo energy. But, when the exchange energy is not equal to the Kondo energy, the presence of disorder might result in the formation of the SG phase.
CeGe is an interesting 4f electron-based system and it undergoes two orderings at $T^I \sim (10.7 \text{ K})$ and $T^\text{II} \sim (4.6 \text{ K})$ at low magnetic field. The former is due to antiferromagnetic ordering while the latter arises due to some spin rearrangement [27]. The high-magnetic-field–dependent studies of physical properties on CeGe revealed the absence of signature of a FL or NFL state [28–31], which is generally observed for other Ce-based compounds [7]. Hence, it would be of interest to investigate the physical properties of CeGe by dilution of the Ce site by a nonmagnetic ion, as there are no such literature reports about these investigations on this compound; to the best of our knowledge. In this letter, we focus on magnetization, thermodynamics and transport properties of Ce$_{0.4}$La$_{0.4}$Ge and Ce$_{0.24}$La$_{0.76}$Ge compounds in the low-temperature region. We choose Ce$_{0.24}$La$_{0.76}$Ge because the ordering temperature of the compound CeGe decrease with the increasing La substitution and it is expected that the magnetic transition temperature may be suppressed below 1.8 K in the Ce$_{0.24}$La$_{0.76}$Ge. Also, the Kondo energy scale may be approximately comparable to the magnetic exchange energy in Ce$_{0.24}$La$_{0.76}$Ge. Moreover, the introduction of La will induce disorder and will also tune the hybridization of localized moments with conduction electrons. This may result in the observation of exotic physical properties. Our results revealed that the magnetic ordering temperature of Ce$_{0.6}$La$_{0.4}$Ge is shifted down in temperature while for Ce$_{0.24}$La$_{0.76}$Ge it is suppressed below 1.8 K. Interestingly, for Ce$_{0.24}$La$_{0.76}$Ge, the variation of heat capacity and resistivity shows the presence ofNFL behavior which arises as a result of the interplay of the disorder effect due to 4f spins (due to the site dilution effect) along with balanced Kondo and magnetic exchange energy. Above 2 tesla, our results divulge the presence of a field-induced magnetic state arising due to the presence of short-range correlations among the magnetic moments. Magnetoresistance scaling indicates that the behavior of the disorder-drivenNFL state is described by the dynamical mean-field theory of the spin glass quantum critical point.

**Experimental.**—High-quality polycrystalline compounds Ce$_{0.6}$La$_{0.4}$Ge (La$_{0.4}$) and Ce$_{0.24}$La$_{0.76}$Ge (La$_{0.76}$) are prepared by arc melting stoichiometric amounts of respective high-purity elements (>99.9%) in an atmosphere of argon. The compounds are re-melted several times to ensure homogeneity and weight losses after final melting is insignificant (<0.5%). X-ray diffraction studies at room temperature established the single-phase nature of these compounds. The indexed XRD is shown in fig. 1(a). The compounds crystallize in orthorhombic structure. For Ce$_{0.6}$La$_{0.4}$Ge, the obtained lattice parameters are $a = 8.398(4) \text{ Å}$, $b = 4.098(6) \text{ Å}$, $c = 6.059(7) \text{ Å}$ and $V = 208.585 \text{ Å}^3$, whereas, for Ce$_{0.24}$La$_{0.76}$Ge the parameters are $a = 8.434(4) \text{ Å}$, $b = 4.116(9) \text{ Å}$, $c = 6.089(7) \text{ Å}$ and $V = 211.454 \text{ Å}^3$. From the lattice parameters it is noted that with an increase in La substitution, the unit cell volume increases.

The peak shifts towards the lower angle side (inset of fig. 1(a)), thereby establishing that the dopant goes to the Ce site. We have also performed the energy dispersive spectroscopy (EDAX) to get the confirmation about the average atomic stoichiometry, which is found to be in accordance with the expected values. Temperature– and magnetic-field–dependent heat capacity and resistivity are performed using the Physical Property Measurement System (PPMS), while temperature-dependent magnetization studies are performed using the Magnetic Property Measurement System (MPMS); both from Quantum design, USA.

**Results and discussions.**—Figure 1(b) shows the temperature-dependent DC susceptibility for La$_{0.4}$ and La$_{0.76}$ compounds at 0.005 tesla under the zero-field–cooling (ZFC) condition. The parent compound CeGe undergoes two antiferromagnetic transitions approximately at 10.7 K and 7.6 K [27]. These transitions are shifted to 4.7 ($T^I$) and 2.7 K ($T^\text{II}$) for La$_{0.4}$ (shown in the inset of fig. 1(b)). For the extreme composition La$_{0.76}$, the transition temperature is suppressed below 1.8 K. The temperature response of the AC susceptibility in the presence of a superimposed DC field of 0.005 tesla for both the compound is shown in fig. 1(c). From the figure, it is noted that the real part of AC susceptibility ($\chi''(\omega)$) for the La$_{0.4}$ compound shows a sharp peak around 4.7 K followed by a weak anomaly around 2.7 K (shown by an arrow) which is in accordance with the transition temperature noted from DC susceptibility. For the La$_{0.76}$ compound, no such peaks are observed. For both compounds, the imaginary part of the AC susceptibility ($\chi''(\omega)$) is insignificant and is not detected by the instrument. Figure 1(d) shows the magnetic-field–dependent isothermal magnetization at the lowest attainable temperature of 1.8 K for both compounds. The magnetization increases with increasing magnetic field; however, at low field, it is observed that the slope of the curve changes with La doping resulting in the increment of the magnetic moment. Also, it is observed that magnetic hysteresis is absent in both of these compounds (not shown).

Figure 2(a) shows the temperature-dependent heat capacity ($C$) for the La$_{0.4}$ and La$_{0.76}$ compounds at 0 tesla. For the La$_{0.4}$ compound, a peak is observed around $T^I$ followed by a weak anomaly around $T^\text{II}$ (shown by an arrow). The curve is featureless for the La$_{0.76}$ compound because the transition temperature is shifted below 1.8 K. For both compounds, it is noted that with decreasing temperature, $C$ increases and attains broad maxima around 155 K. Such feature possibly arises due to the presence of incoherent Kondo scattering along with the CEF effect [32]. The upper inset of fig. 2(a) shows the heat capacity divided by temperature as a function of $T^2$. To subtract the phonon contribution, we fit the equation: $C/T = \gamma + \beta T^2$ in the temperature range of 10–30 K. In this equation, $\gamma$ and $\beta$ are the electronic and phonon contributions of the heat capacity. The obtained
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values of $\gamma$ are 94.2 and 2.4 mJ/mol K$^2$ for the La$_{0.4}$ and La$_{0.76}$ compounds, respectively. It is noted that $\gamma$ decreases with increasing La substitution resulting in the decrease of the screening effect of magnetic moments. This effect is due to the fact that the increase in unit cell volume with increasing La substitution results in the decrement of $Jn(E_f)$ (where $J$ is the exchange interaction parameter of the $f$-$d$ hybridization and $n(E_f)$ are the electronic states per unit volume at Fermi level) [33]. The lower inset of fig. 2(a) shows the temperature-dependent $C'/T$ for the La$_{0.76}$ compound, which is obtained after subtracting the phonon contribution from heat capacity up to the lowest temperature using the equation: $C'/T = \frac{\gamma_0 - \alpha T^b}{T}$. The curve increases with decreasing temperature and it is fitted (in the temperature range 1.8–3.5 K) using the equation $C'/T = \frac{\gamma_0 - \alpha T^b}{T}$, where $b$ is temperature exponent, $\gamma_0$ and $\alpha$ are the free fitting parameters [7]. Best fitting is obtained for $b = 0.5 \pm 0.03$, $\gamma_0$ and $\alpha$ are $\sim 0.890 \pm 0.01$ and $0.45 \pm 0.01$, respectively. The value of $b$ indicates the presence of NFL behavior in the low-temperature regime. This feature might arise due to a critical spin fluctuation because of the equivalent strength of the Kondo and magnetic exchange energy [25]. Figure 2(b) shows the temperature-dependent normalized resistivity ($\rho/\rho_{200}$) for both compounds at 0 tesla. With decreasing temperature, resistivity decreases which is a characteristic of the metallic behavior. However, for the La$_{0.4}$ compound, a nonlinear deviation is noted below 160 K. Such feature can possibly arise due to the effect of partial screening of magnetic moments via conduction
and thermodynamic properties in the low-temperature region [32,34]. For the La$_{0.4}$ compound, it is noted that resistivity rises around $T^4$, followed by a maximum around $T^{11}$ (upper inset of fig. 2(b)). A similar effect has been reported for CeGe which undergoes a gap opening at $T^1$ and some spin rearrangement at $T^{11}$ [27]. Below $T^{11}$, coherence due to some spin rearrangement may arise, which results in a decrease in resistivity. For the La$_{0.76}$ compound, no such upturn is noted. Hence, it can be said that with La-substitution gap opening is shifted to lower temperature (below 1.8 K) or is suppressed. For the La$_{0.76}$ compound, the variation of resistivity in the temperature range from 1.8 to 3.5 K is fitted with the following equation: \[ \rho = \rho_0 + AT^n, \]
where $\rho_0$ is the residual resistivity, $A$ is associated with the scattering of conduction electrons and $\alpha$ is the temperature exponent [24]. Best fitting is obtained for $n = 1.6 \pm 0.02$. The lower inset of fig. 2(b) shows $\rho$ plotted as a function of $T^{1.6}$ which illustrates a linear behavior (the arrow indicates the deviation in the curve around 3.5 K). The value of the exponent $n$ also indicates the observation of the NFL behavior in this compound. As per literature reports, $\rho$ varies with $T^d$ due to critical spin fluctuations [5,6]. However, the observation of an unusual variation of $\rho$ (varies with $T^{1.6}$) suggests that the observed NFL behavior could be caused by the interplay of the disorder effect due to 4f spins (due to the site dilution effect) along with critical spin fluctuations. A similar effect has been reported for other compounds like CeNi$_2$Ge$_2$ and CeCu$_2$Si$_2$ [35]. Hence, it can be said that with increasing La substitution the hybridization of magnetic moments with the conduction electrons is modified resulting in the observation of an unusual NFL behavior in the La$_{0.76}$ compound.

In order to further explore this behavior, we have studied the temperature response of heat capacity and resistivity under the influence of magnetic field up to 14 tesla.

Figure 3(a) shows the temperature-dependent $C'/T$ under different magnetic fields. As the magnetic field increases up to 2 tesla, the nature of the slope of $C'/T$ remains unchanged, indicating the persistence of the NFL behavior. As the magnetic field is increased further, a curvature is noted around 2.2 K for 3 tesla which indicates a new magnetic state due to the dominance of short-range correlations (SRC) among the localized magnetic moments, which results in the suppression of NFL behavior. Above 3 tesla, this curvature becomes more prominent with increasing magnetic field. The weak signature of the curvature is also noted in the temperature response of the $M/H$ curves at high fields (inset of fig. 3(a)). The temperature-dependent entropy ($S'$) curve at 0 tesla is shown in the inset of fig. 3(b). The $S'$ decreases with decreasing temperature and the obtained entropy is about 0.30 (0.052 R ln2) around 3.5 K. The observed entropy is very small compared to the compound which shows the multi-channel Kondo effect [36]. Hence, it can be said that the existence of adequate disorder by the Ce-site dilution might be responsible for such low

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entropy [36]. The observation of NFL behavior is further analyzed by the scaling of $C'/T$ with the characteristic temperature $T^*$, which depends upon the strength of the Kondo energy [11,35]. Figure 3(b) shows the scaled $C'/T$ vs. $T/T^*$ at different fields (0–2 T). $T^*$ is the characteristic temperature and is taken as 1.8, 2, 2.2 and 2.6 K for $H = 0, 0.5, 1$ and 2 tesla, respectively. Inset: $T$-dependent entropy ($S'$) at 0 tesla.

Figure 4(a) shows the temperature-dependent resistivity $\rho$ under different fields. The resistivity decreases linearly with decreasing temperature for all fields due to the metallic behavior of the compounds. In the low-temperature regime, the curves are fitted with the equation $\rho = \rho_0 + AT^n$ (not shown). The inset of fig. 4(a) shows the magnetic-field–dependent residual resistivity ($\rho_0$). It is observed that $\rho_0$ increases up to 1 tesla (approximately) due to increasing disorder. At high fields, $\rho_0$ decreases remarkably. It might be due to the dominance of SRC because of the presence of strong exchange coupling among

Fig. 3: (a) $T$-dependent heat capacity $C'/T$ in field range 0–14 tesla for the La$_{0.76}$ compound. Inset: $T$ (logarithmic scale)-dependent DC susceptibility ($M/H$) at high fields. (b) $C'/T$ vs. $T/T^*$ (logarithmic scale) at different fields (0–2 T). $T^*$ is the characteristic temperature and is taken as 1.8, 2, 2.2 and 2.6 K for $H = 0, 0.5, 1$ and 2 tesla, respectively. Inset: $T$-dependent entropy ($S'$) at 0 tesla.

Fig. 4: (a) $T$-dependent resistivity in field range 0–14 tesla for the La$_{0.76}$ compound. Inset: magnetic-field–dependent residual resistivity ($\rho_0$). (b) Scaling plots of the resistivity in the low-temperature region and field range 0–1 tesla. Upper inset: $T$-dependent $\rho - \rho_0$ in the field range 0–1 tesla. Lower inset: $T$-dependent $M/H$ at 1 tesla. The red line shows the fitting using the equation $\chi = \chi(0) - cT^\beta$. 

Figure 4(a) shows the temperature-dependent resistivity $\rho$ under different fields. The resistivity decreases linearly with decreasing temperature for all fields due to the metallic behavior of the compounds. In the low-temperature regime, the curves are fitted with the equation $\rho = \rho_0 + AT^n$ (not shown). The inset of fig. 4(a) shows the magnetic-field–dependent residual resistivity ($\rho_0$). It is observed that $\rho_0$ increases up to 1 tesla (approximately) due to increasing disorder. At high fields, $\rho_0$ decreases remarkably. It might be due to the dominance of SRC because of the presence of strong exchange coupling among
the localized magnetic moments. It is also to be noted that the obtained value of the exponent \( n \) is quite away from 2 (the value of \( n \) for the FL state) at high fields. Hence, it can be said that the dominance of the exchange energy impedes the recovery of the FL state, which is also observed in other Ce-based compounds [24]. Possibly the development of SRC among the spins impedes the recovery of the FL state. To probe the exact nature of this magnetic state, low-temperature neutron diffraction measurements are warranted, which is beyond the scope of this manuscript. Interestingly, as per literature reports, disorder-driven NFL state is analyzed by the dynamical mean-field theory of the spin glass quantum critical point (SG QCP) [24]. In the absence of disorder, the dynamical mean-field theory describes the FL behavior in the low-temperature region. However, the presence of significant disorder results in the inhomogeneous scattering which affects the electrical transport properties and observation of NFL behavior [23]. Hence, in our case, the presence of disorder along with coupling of the conduction electron with critical spin fluctuations is described by SG QCP, which has been predicted to understand such anomalous variation of resistivity. Also, it has been reported that the physical properties depend on the effective distance to the QCP (\( \Delta(T,H) \)) and the resistivity in the form of scaling: \( \rho(T,H) - \rho(0,0) \propto t^{3/2} \chi(t/\Delta) \), where \( t = T/T_0 \) (\( T_0 \) is the temperature which depends upon \( J \)) and \( \Psi(x) \) is the scaling function [24]. For disorder-driven NFL, the equation in the preceding line describes the \( T^{3/2} \)-dependence of \( \rho \). \( \Delta \) is described as \( \Delta = \Delta_0 + 2(\Delta_0)^{1/2}/t\{[1 + t/2(3)^{1/2}\Delta_0]^{1/2} - 1\} \), where \( \Delta_0 = r + (H/H_0)^2 \). \( r(=1 - J/J_c) \) is the measure of the distance to the realistic QCP and \( H_0 = J_c/(g\mu_B) \) (\( g \) is the gyromagnetic ratio of the Ce ion, taken as 2 and \( \mu_B \) is the Bohr magnetron). For the critical coupling (at the QCP), the exchange energy \( J \) becomes \( J_c \) (critical exchange energy) and \( \Delta_0 \) approximately equals \( (H/H_0)^2 \). The susceptibility (\( \chi = M/H \)) for the critical coupling has the NFL behavior and is described as \( \chi = \chi(0) - cT^\beta \), for \( T/T_0 \leq 1 \), where \( \chi(0) = (g\mu_B^2)/J_c \) is the zero-temperature susceptibility and \( c = (g\mu_B^2)/J_c (T_0)^{0.75} \) is a coefficient [26]. A best fitting is obtained for \( \beta = 0.75 \pm 0.01 \). The inset of the fig. 4(b) shows the linear dependence of \( M/H \) with \( T^{0.75} - 1 \) tesla. The obtained values \( J_c \) and \( T_0 \) are approximately 10.2 K and 7 K, respectively. Also, from the equation \( \chi = \chi(0) - cT^\beta \), we estimate the characteristic field \( H_0 = \mu_B/\chi(0) \) which is near to 7.6 tesla. The upper inset of fig. 4(b) shows the temperature-dependent \( \rho(T,H) - \rho(0,0) \) curves at magnetic fields of 0–1 tesla. From these curves the best scaling of \( \log[\rho(T,H) - \rho(0,0)]/t^{1.5} \) vs. \( \log(t/\Delta) \) is obtained for the \( T_0 = 7 \) K, \( H_0 = 9.6 \) tesla (we got a better scaling for 9.6 tesla instead of 7.6 tesla) and \( r = 0.385 \) (fig. 4(b)). A similar form of scaling has been reported in Ce\((Ru_{0.5}Rh_{0.5})_2Si_2\), which also shows the disorder-driven NFL behavior [24]. The scaling of the resistivity indicates that the behavior of this compound is described by the dynamical mean-field theory of SG QCP. Also, we calculate the value of \( \delta J = (J_c - J) = rJ_c = 3.9 \) K. The \( \delta J \) determined from scaling is close to the theoretical estimated value in ref. [26]. It has been reported that for \( J \leq J_c \), there are two possibilities with decreasing temperature: FL behavior or a magnetic state, depending on the factor \( Jn(E_f) \) [33]. For the La\(_{0.76}\) compound, the FL behavior has been ruled out and our results indicate that the field induced a new magnetic state which arises due to the presence of SRC among the magnetic moments. A similar type of behavior is also reported in ref. [37]. Above 2 tesla, NFL behavior is suppressed and these weak interactions strengthen, leading to the growth of SRC among spin at high fields. This SRC among the spins results in the development of a new magnetic state at high fields. Thus, it can be said that in La\(_{0.76}\) compound, the NFL phase is below 3.5 K and around 2 tesla. In this phase \( M/H, C'/T \) and \( \rho \) vary with \( T^{0.75}, T^{0.5} \) and \( T^{1.6} \), respectively. The behavior of the NFL phase has been described by the dynamical mean-field theory of SG QCP. The phase above 2 tesla and below the temperature where a new anomaly is observed in the heat capacity curve for the respective field is a new magnetic phase. In this phase, SRC among spins is prevalent due to the dominance of the interaction between the localized magnetic moments via conduction electrons because of the presence of a strong exchange coupling. The temperature of the anomaly increases with increasing magnetic field due to an increase of the coupling strength.

Summary. — In this work, we have studied the low-temperature magnetic, thermodynamic and transport properties of the La\(_{0.4}\) and La\(_{0.76}\) compounds. It is observed that with increasing La substitution, the ordering temperature is shifted down in temperature for La\(_{0.4}\) (as compared to CeGe) and is suppressed below 1.8 K for La\(_{0.76}\). Interestingly, the temperature variation of resistivity and heat capacity suggests the presence of an unusual NFL behavior which arises due to the presence of disorder. As the magnetic field is increased above 2 tesla, the field induced a new magnetic state is noted which arises due to the presence of the strong short-range correlations among the magnetic moments. The magnetoresistance scaling indicates that the behavior of the disorder-driven NFL behavior is described by the dynamical mean-field theory of the spin glass quantum critical point.

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