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Evolution of physical properties in the $Ce_{1-x}La_xFeGe_3$ series: crossover from Kondo lattice to single ion Kondo impurity systems

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Abstract. Evolution of the structural, magnetic, transport and magneto-transport properties of the Ce_{1-x}La_xFeGe₃ series is studied by powder x-ray diffraction, ⁵⁷Fe Mössbauer spectroscopy, magnetization and resistivity in zero and applied magnetic field. Changes from concentrated Kondo system-like ($x \le 0.3$) to single Kondo ion-like ($x \ge 0.65$) behaviour are observed. Magneto-resistivity behaviour in both regimes is discussed within the existing theoretical views which include non-negligible 'normal' metal contributions.

1. Introduction

Recently, a new moderately heavy fermion material CeFeGe₃ ($\gamma \approx 150 \text{ mJ mol}^{-1} \text{ K}^{-2}$), with a high Kondo temperature ($T_K \approx 100$ K) and well defined integer valence of Ce (Ce³⁺) at high temperatures, was reported [1,2]. This compound is one of the few rare earth heavy fermion compounds containing Fe. It crystallizes in a tetragonal BaNiSn₃-type structure (figure 1) [3] which is closely related to $ThCr_2Si_2$ -type structure, where a number of heavy fermion compounds as well as exotic superconductors and complex magnetic materials were reported [4]. While CeFeGe₃ can be characterized as a concentrated Kondo system [1, 2], LaFeGe₃ is a 'normal metal' with no anomalies in transport or magnetic properties. Some compounds from the $Ce_{1-x}La_xFeGe_3$ series were recently studied by means of magnetic susceptibility, zero-field resistivity and specific heat measurements [5,6]. In this work we present a detailed study of the evolution of the structure, ⁵⁷Fe Mössbauer measurements, lowtemperature magnetization and magneto-transport properties of the $Ce_{1-x}La_xFeGe_3$ series paying particular attention to its resistivity in zero and applied magnetic fields. It has to be noted that, in general terms, we expect to observe the evolution from the concentrated Kondo system, for high Ce concentrations, to the single ion Kondo impurity behaviour, in low Ce limit, and finally to normal metal. The details of this evolution, like the position of the crossover concentration region between the concentrated Kondo system regime and the single ion Kondo impurity behaviour, as well as the absence or presence of the magnetic instability for the intermediate La concentrations, are dependent on the series under study. The details of the evolution of the ground state are addressed in the present work.

Evolution of the strongly correlated electron behaviour in the $Ce_{1-x}La_xFeGe_3$ series, as well as possible magnetic instabilities, usually unambiguously affect the temperaturedependent resistivity. Theoretical predictions for the magneto-resistance behaviour both in

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Figure 1. Crystalline structure of the $Ce_{1-x}La_xFeGe_3$ series [3].

the single Kondo impurity regime and for the concentrated Kondo systems exist in the literature [7,8] therefore, temperature-dependent resistivity and field-dependent magneto-resistance were chosen as the most appropriate techniques for this study.

2. Experimental

The polycrystalline samples of the Ce_{1-x}La_xFeGe₃ series with x = 0, 0.1, 0.3, 0.5, 0.65, 0.7, 0.75, 0.9 and 1 were synthesized by conventional arc-melting of the stoichiometric amounts of high purity elements in a partial (~200 Torr) Ar atmosphere with subsequent annealing at ~750 °C for 5 days. Room temperature powder x-ray diffraction patterns taken after annealing do not contain any impurity lines. The lattice parameters were fitted using a Rietveld refinement program [9]. Magnetization was measured using a commercial Quantum Design MPMS-5 SQUID magnetometer in Ames Laboratory/Iowa State University. Temperature- and field-dependent resistivity was measured on thin rectangular slabs by conventional four probe techniques using a Linear Research LR-700 AC resistance bridge (frequency f = 16 Hz, current density $j \approx 0.3$ A cm⁻²). ⁵⁷Fe Mössbauer spectroscopy (MS) measurements were performed at 300 K and 4.2 K in a transmission geometry and constant acceleration mode with a ⁵⁷Co in Rh matrix Mössbauer source. The source and absorber were kept at the same temperature for all measurements. The obtained isomer shift (IS) values are relative to the α -Fe absorber.

It has to be mentioned that magnetization M(H) measurements at low temperatures reveal very small, less than 1%, (undetectable by x-ray diffraction) amounts of magnetically ordered impurities, which saturate in applied fields of about 10–15 kG (the existence of similar levels of impurities has already been mentioned in [1, 2, 5, 6]). Due to these impurities, the M(T) measurements in a constant field inevitably have non-negligible spurious contribution. To eliminate, at least partially, this impurity signal the M(H) measurements were performed at a number of temperatures and the results presented later are M/H values obtained from the close-to-linear portion (H = 25-55 kG) of the M(H) curves taken at constant temperature.

Polycrystalline samples usually suffer several problems which are important in the evaluation of the *absolute* values of resistivity: (a) voids and micro-cracks; (b) the possibility of (uncontrolled) preferential orientation of the crystallites within the sample due to the thermal gradient at the time of its formation. The rectangular bars for the transport measurements were cut in such a way that, for all samples, the electric current during the measurement was flowing in approximately the same direction with respect to the thermal gradient which existed during solidification of the samples in the arc-furnace. Care was taken to choose, for the transport measurements, samples without visible cracks (as inspected by optical microscope). These precautions allowed us to observe semi-quantitative trends in resistivity through the series (see later).

3. Results

Room-temperature powder x-ray diffraction measurements confirm that the samples have single phase BaNiSn₃-type structure. The variation of the lattice parameters and the unit cell volume follows Vegard law. From CeFeGe₃ to LaFeGe₃ the lattice parameters a and c increase linearly with changes of 0.8% and 0.3%, respectively. The unit cell volume changes are about 2% (figure 2), the a/c ratio increases slightly from 0.435 for CeFeGe₃ to 0.437 for LaFeGe₃. These changes in the unit cell dimensions are consistent with the data reported in [1, 2, 5, 6].

Figure 3 shows the representative Mössbauer spectra at 4.2 K for the $Ce_{1-x}La_xFeGe_3$ series, which are similar to the ones obtained at 300 K. All the spectra could be analysed with one symmetric unresolved doublet with line width, $W \approx 0.25$ mm s⁻¹, and quadrupole splitting (QS) around 0.22 mm s⁻¹. This speaks for a unique ⁵⁷Fe site in agreement with the crystallographic position in the BaNiSn₃-type structure. The sharp line width together with the existence of a single Fe site is indicative of good sample quality, as is also suggested by x-ray diffraction measurements. The values of the isomer shift and quadrupole splitting, obtained from fits of the Mössbauer spectra at two different temperatures, are shown in figure 4. Both parameters vary linearly with La concentration. The slopes of IS against *x*-La lines are basically the same, while QS at T = 4.2 K is changing somewhat faster with *x*-La than at T = 300 K.

Low-temperature magnetic susceptibility as a function of La concentration is shown in figure 5. Magnetic susceptibility per formula unit decreases with increase of x-La, which is expected since magnetic Ce is replaced by non-magnetic La, however, this dependence is not linear and the susceptibility per Ce has a peak around x = 0.5 (figure 5, inset). Temperature-dependent zero-field electrical resistivity for representative samples is shown in figure 6. The curve for LaFeGe₃ has a typical 'normal' metallic behaviour with a quite large linear high-temperature region dominated by electron-phonon scattering starting at about 100 K, high residual resistance ratio (RRR ≈ 50) and rather low room temperature resistivity ($\rho(300 \text{ K}) \approx 90 \ \mu\Omega$ cm). The temperature-dependent resistivity of CeFeGe₃ is typical for a concentrated Kondo system (CKS) with a high Kondo temperature: at high temperatures (down to 150–100 K) $\rho(T)$ slightly decreases with the decrease of temperature, then a much steeper decrease occurs which is followed by an inflection point and approximately



Figure 2. Lattice parameters and unit cell volume in the $Ce_{1-x}La_xFeGe_3$ series.

 T^2 behaviour at low temperatures. The residual resistance ratio for CeFeGe₃ is about 45. It has to be noted that for our CeFeGe3 and LaFeGe3 samples, the RRR values are slightly higher than those reported in [1, 2] which may indicate a somewhat superior sample quality. The residual resistance ratios decrease rapidly with small La or Ce alloying in the pure end compounds (figure 7, inset). The absolute values of room temperature and residual (at T = 2 K) resistivity, as a function of La concentration (figure 7), follow expected trends: the room temperature resistivity monotonically decreases with increases of x-La, since the additional contribution of the scattering on the paramagnetic Ce ions decreases, and the residual resistivity behaviour is close to the parabola expected from Nordheim rule. For three samples with La concentration close to x = 0.7, the points on the graph deviate from the trend observed for the other ones, however, these three samples have no anomalies in other properties. Therefore, it is tempting to assume that the density of voids and microcracks in the samples for this region of x-La is higher (for presently unknown reasons). That leads to much higher errors in the estimation of the effective cross section of the samples. Indeed, if we scale the room temperature resistivity values to the expected curve, the residual resistivity values will be shifted to their exact places on Nordheim rule parabola (figure 7). For intermediate La concentrations, a gradual change of the $\rho(T)$ curve, from



Figure 3. Representative ⁵⁷Fe Mössbauer spectra of the Ce_{1-x}La_xFeGe₃ series at T = 4.2 K.

CKS-like behaviour of resistivity for low x-La, to single Kondo impurity-like shape for high x-La, is observed (figure 6). For x = 0.5 a sort of crossover between these two regimes is noticed: the inflection point at $T \approx 55$ K, remnant of CKS-like behaviour, is followed by the resistivity minimum around 30 K and further increases of the resistivity at lower temperatures, characteristic for single Kondo impurity regimes. The position of the inflection points, separately for low x-La and high x-La, which are related, respectively, to the values of the coherence temperature and single impurity Kondo temperature, as well as the position of the resistivity minimum for high x-La, are quite insensitive to La concentration (figure 8), except for x = 0.5, where a 'mixture' of both regimes occurs.

Since the Kondo and coherence temperatures of CeFeGe₃ are high [1, 2], the Fermi liquid regime characterized by T^2 dependence of resistivity occurs in a rather large temperature range. The low temperature T^2 behaviour is clearly seen also for x = 0.1 and 0.3 (figure 9). The coefficient A of this term decreases with the increase of x-La (figure 9, inset). The samples close to the other end of the series (high La concentrations) show (figure 6) a



Figure 4. Isomer shift and quadrupole splitting in the $Ce_{1-{\it x}}La_{\it x}FeGe_3$ series at 300 K and 4.2 K.



Figure 5. Low-temperature magnetic susceptibility in the $Ce_{1-x}La_xFeGe_3$ series per formula unit and (inset) per Ce. Lines are guides for the eye. Approximate error bars for T = 2 K are plotted in the inset.

Kondo minimum at about 40–50 K and a significant increase of resistivity with decreasing temperature followed by the tendency to saturate at temperatures close to 2 K. This type of behaviour is observed up to rather high (35%) concentrations of Ce, that points to rather small inter-impurity correlations which are usually much weaker in rare earth alloys than,



Figure 6. Representative zero-field resistivity curves for the $Ce_{1-x}La_xFeGe_3$ series.

for example, in 3d systems. Zero-field resistivity in low Ce concentration regimes can be analysed in the same way as was done, for example, for the (**La**, Ce)B₆ system [10]. A plot of the resistivity at the Kondo minimum against Ce concentration gives a straight line over a wide range of Ce concentrations (figure 10) with an extrapolated resistivity value of about 3.3 $\mu\Omega$ cm at zero Ce concentration, which agrees quite well with the residual resistivity of the pure LaFeGe₃ sample. This suggests that for low Ce concentrations the samples have roughly the same residual resistivity from scattering by non-magnetic impurities (contain the same amount of structural disorder). Subtracting the residual resistivity of the pure LaFeGe₃ sample from the low-temperature (T = 1.6 K, approaching saturation) resistivity values for the Ce_{1-x}La_xFeGe₃ series with low Ce concentrations, we can estimate the unitary limit $\rho_u \approx 3.6 \ \mu\Omega$ cm at.%⁻¹ Ce (figure 10, inset). For Ce concentrations between 10% and 35% changes in the obtained unitary limit are small, probably indicating weak inter-impurity correlations.

Representative curves for transverse magneto-resistance measured for the $Ce_{1-x}La_xFeGe_3$ series at different temperatures, in applied fields up to 80 kG, are shown in figure 11. For



Figure 7. Room temperature and residual resistivities in the $Ce_{1-x}La_xFeGe_3$ series. Open circles and stars, raw data; closed and crossed circles, scaled with room temperature resistivities (see text). Inset: residual resistance ratio as a function of La concentration.



Figure 8. Position of Kondo minimum and inflection points in $\rho(T)$ curves as a function of La concentration in the Ce_{1-x}La_xFeGe₃ series. Shadowed area, crossover region.

all samples the magneto-resistance at 1.6 K and 4.2 K is positive. For pure CeFeGe₃ and LaFeGe₃ the relative change of the resistivity in the applied field is high (at T = 1.6 K, $\Delta \rho(H)/\rho(0)$ it is about 20% and 50%, respectively, at 80 kG). At higher temperatures the magneto-resistance decreases. The relative change of resistivity $\Delta \rho(80 \text{ kG})/\rho(0)$ also



Figure 9. $\rho(T)-\rho_{LaFeGe_3}(T)$ curves for several La concentrations as a function of T^2 . Inset: T^2 coefficients as a function of x-La.

decreases for intermediate La concentrations. The relative change of resistivity in the applied field of 80 kG at two different temperatures is plotted as a function of La concentration in figure 12. It has to be noted that since zero-field low-temperature resistivity varies by more than an order of magnitude for samples of the Ce_{1-x}La_xFeGe₃ series presentation of the magneto-resistance data as a *relative* change of resistivity in an applied field may hide some information due to large changes in the denominator of the ratio $\Delta \rho (80 \text{ kG}) / \rho (0)$. An alternative plot of the *absolute* changes of resistivity by application of the field of 80 kG as a function of *x*-La is presented in the inset to figure 12. This plot shows an anomaly in $\Delta \rho (80 \text{ kG})$ for *x*-La around 0.6.

Evolution of the $\Delta \rho(H)/\rho(0)$ curves with x-La is rather peculiar (figure 11). The magneto-resistance remains positive for $x \ge 0.5$ in the whole field range up to 80 kG and exhibits more complicated behaviour for x < 0.5. In the latter region of lower La concentrations at temperatures higher than approximately 30 K the magneto-resistance first turns negative, then reaches a minimum and returns positive at higher fields. Similar high-temperature behaviour is observed for pure CeFeGe₃.

4. Discussion

The variation of unit cell parameters can be easily understood from 'geometric' considerations: the ionic radius of La^{3+} is larger than that of Ce^{3+} which causes an increase of the lattice parameters and the unit cell volume with an increase of *x*-La.

⁵⁷Fe Mössbauer measurements in the Ce_{1-x}La_xFeGe₃ series indicate that, as well as in compounds with 'parent' ThCr₂Si₂ structure [11, 12], Fe atoms do not carry a magnetic



Figure 10. Low-temperature saturation resistivity and resistivity at Kondo minimum as a function of Ce concentration. Inset: estimated unitary resistivity limit (the line is a guide for the eye).

moment (within the accuracy of the technique, $\mu_{Fe} < 0.05 \ \mu_B$). Moreover, no hyperfine field, transferred from the 4f moments of Ce, was observed at the Fe site down to 4.2 K. Fe atoms are located on the top of a pyramid the base of which is formed by four Ge atoms in Ge2 position (figure 1), leading to a lattice contribution to the quadrupole interaction. The quadrupole splitting varies linearly with La concentration (figure 4) with different negative linear coefficients for T = 300 K and T = 4.2 K. Expansion of the lattice with La substitution occurs both in the (ab) plane and along the c axis, with increasing a/c ratio. Consequently, the Ge2-Fe-Ge2 bond angles increase, which probably results in a reduction of the lattice contribution to the quadrupole interaction. For the Ce-rich side the difference between the QS values measured at 4.2 K and 300 K is larger than for the La-rich side. This may be due to the fact that the population of the crystalline electric field (CEF) levels at fixed temperature is, possibly, concentration dependent [13]. The isomer shift increases linearly with La concentration with similar slopes at 4.2 K and 300 K with room temperature values being slightly higher (figure 4). Regarding the IS changes with La concentration, for a ⁵⁷Fe:Fe matrix, the volume dependence of the isomer shift was estimated [14] as $\Delta IS/(\Delta V/V) = 1.37-1.40$ mm s⁻¹. The variation of the isomer shift in Ce_{1-x}La_xFeGe₃ compounds obtained from our measurements at 300 K and 4.2 K are $\Delta IS = 0.0146$ mm s⁻¹ and 0.0193 mm s⁻¹, respectively. Thus, the estimated volume change is $\Delta V/V \approx 1\%$ at 300 K and $\Delta V/V \approx 1.4\%$ at 4.2 K. These values are in good agreement with the unit cell volume change ($\Delta V/V \approx 2\%$) within the series, obtained from room temperature x-ray diffraction measurements. Therefore, variation of the isomer shift at fixed temperature can be attributed mainly to changes of the unit cell volume with La concentration: as the lattice expands, the density of s-electrons at the Fe nuclei decreases, implying an increase of the isomer shift for higher La concentrations.



Figure 11. Representative magneto-resistance curves for the $Ce_{1-x}La_xFeGe_3$ series. Note, due to the difference in magneto-resistance, results for CeFeGe₃ are presented on two different panels.

Pure CeFeGe₃ was classified as a concentrated Kondo system. Zero-field resistivity measurements suggest that the same may be true for $Ce_{1-x}La_xFeGe_3$ compounds with x < x0.5. For heavy fermion compounds, the following relations between γ the linear coefficient in the temperature dependence of the specific heat, $\chi(0)$ the zero-temperature magnetic susceptibility and A the coefficient of the T^2 term in resistivity are expected [15]: $\chi(0) \propto \gamma$ and $A \propto \gamma^2$. Measurements of $\chi(2 \text{ K})$ and A for x < 0.5 in this work together with the data for γ from [5, 6] are, at least, in qualitative agreement with the expected behaviour. The fact that low-temperature magnetic susceptibility (as well as γ) per formula unit decreases with the increase of x-La and at the same time the value per Ce increases supposedly reflects the balance of two opposite trends: La (electron configuration [Xe] $5d^{1}6s^{2}$) substitution leads to a decrease in the density of 'heavy' f-electrons, while negative chemical pressure according to Doniach phase diagram [16] or its version for CKS [17, 18] increases the mass of the Ce f-electrons trying to bring the system closer to the critical point on the phase diagram. For the studied $Ce_{1-x}La_xFeGe_3$ series, the pure CeFeGe₃ compound is far from the critical point (high Kondo and coherence temperatures) and the change in the lattice parameters with x-La seems not to be large enough to reach the critical point and then enter the magnetic ground state, as was observed, for example, in the $Ce_{1-x}La_xRu_2Si_2$ system [19].



Figure 12. Relative change of resistivity at 1.6 K and 60 K in an applied field of 80 kG in the $Ce_{1-x}La_xFeGe_3$ series. Inset, absolute change of resistivity at 4.2 K in an applied field of 80 kG in the $Ce_{1-x}La_xFeGe_3$ series. Open circles, raw data; closed circles, scaled with room-temperature resistivity (see text above).

Several experimental observations are noted: the peak in low-temperature magnetic susceptibility per Ce; the anomaly in the absolute change of the resistivity in the applied field; and finally the qualitative changes in the shape of the zero-field $\rho(T)$ curves suggest that a crossover between two different regimes, CKS and single impurity Kondo effect, occurs for values of *x*-La around 0.5.

It has to be noted that single Kondo impurity-like behaviour in the Ce_{1-x}La_xFeGe₃ system holds up to $x \leq 0.65$, a rather high ($\geq 35\%$) concentration of Ce, i.e. contributions from the independent Kondo centres is dominant over a wide range of concentrations. Similar behaviour was observed, for example, in Ce_xLa_{1-x}Cu₆ [20] and Ce_xLa_{1-x}Pb₃ [21] systems with single Kondo impurity-like behaviour dominating up to as high as 80% Ce concentration. This behaviour is different, for example, from intermetallic compounds containing U, where the single Kondo impurity regime occurs only for U concentrations $x \ll 1$. The theoretical treatment of this difference between intermetallic Kondo systems containing Ce and U, using Nozieres approach in Fermi liquid theory for magnetic impurities as scattering centres at low ($T < T_K$) temperatures, was presented by Gor'kov and Kim [22]. They suggest that compounds with Ce are much closer to the unitary limit than ones with U, and that in formation of the collective ground state, the change in the density of states is the dominant effect in a Kondo lattice, where the single-centre phase shift is close to the unitary limit, while the magnetic (RKKY) interactions, between local impurity centres, become important in a system where the phase shift is different from $\pi/2$ (compounds with U).

For low Ce concentrations (single Kondo impurity-like behaviour) the unitary limit of resistivity at low temperatures is caused by saturation of the Kondo divergence due to complete scattering of partial waves from magnetic impurities. It can be written as [23]

$$\rho_u = 2(l+1)2m^*c/[\pi\hbar z e^2 N(E_F)]$$

where m^* is the effective mass, z is the number of conduction electrons per host atom, $N(E_F)$ is the density of states for one spin direction and l is the orbital quantum number of the valence electron of magnetic impurity for the f-electron of Ce, l = 3. Since no band structure calculations and/or Fermi surface parameters measurements for LaFeGe₃ were reported up to now, it is impossible to compare experimental estimations for the unitary limit of resistivity with theoretical predictions. The obtained experimental values are of the same order of magnitude as those reported for (La, Ce)B₆ samples [10], and so they are probably physically reasonable.

In the simple case of a compound in the single impurity Kondo regime, the expected sign of magneto-resistance is negative [7]. This is different from our results for x > 0.5for the $Ce_{1-x}La_xFeGe_3$ system. To understand this discrepancy, it must be remembered, that for pure LaFeGe₃ the magneto-resistance is high and positive. For normal metals the magneto-resistance is positive and its magnitude is governed by the product $\omega \tau$, where ω is a cyclotron frequency and τ is a mean free time. Since the residual resistivity for LaFeGe₃ is low, the mean free time and subsequently the low-temperature magneto-resistance is high. In the presence of magnetic impurities, in low and intermediate field limits ($\omega \tau \leq 1$), the magnetic field causes bending of the electron orbits which leads to a positive contribution to magneto-resistance $\Delta \rho_1 \propto \rho(\omega \tau)^{\alpha}$, $\alpha \approx 2$, (the dominating contribution in normal metals in low and intermediate field limits) and a negative contribution associated with spin polarization of the impurity atoms $\Delta \rho_2 \propto -\rho_J^{(0)} (g S \mu_B H/T)^2$ (for $S \mu_B H \ll T$) where μ_B is the Bohr magneton and $gS\mu_B$ is the magnetic moment of impurity [24]. This means that the total change in resistivity in an applied field may be either positive or negative. In the case of the Ce_{1-x}La_xFeGe₃ system (x > 0.5) the first term is supposedly dominant and the resultant magneto-resistance is positive. For the CKS regime the theory [8] predicts positive magneto-resistance for $T \ll T_K$ which is consistent with our 1.6 K and 4.2 K results, since the estimation of T_K for pure CeFeGe₃ is about 100 K [1,2]. However, for $T \ge T_K$ magneto-resistance is predicted [8] to turn negative. This seems to be in agreement with the experimental results for x < 0.5, if we suppose that even in the CKS limit some 'normal' (positive) contribution of magneto-resistance is present which turns the total magneto-resistance positive at high fields.

5. Conclusions

The substitution of Ce by La in the Ce_{1-x}La_xFeGe₃ series causes a linear increase of lattice parameters in agreement with Vegard law. Zero-field resistance reveals a gradual change from concentrated Kondo system-like behaviour at low x-La to single Kondo impurity-like behaviour for high x-La with the crossover between these two regimes at $x \approx 0.5$. In the whole series no traces of magnetic instability were observed down to 1.6 K, which suggests that negative chemical pressure induced by La substitution in the series is not strong enough to overcome Kondo screening of the Ce moments and dilution of magnetic Ce ions by nonmagnetic La ones. In the single Kondo impurity regime the unitary resistivity limit is estimated to be about 3.6 $\mu\Omega$ cm at.%⁻¹ Ce. In the concentrated Kondo system regime, $\rho = AT^2$ (Fermi liquid), resistivity behaviour at low temperature was observed up to x = 0.3 with the coefficient A decreasing with an increase of x-La. The peculiarities of the magneto-resistance behaviour in these two regimes were explained by the balance between a 'normal' metal (positive) contribution and Kondo behaviour. According to Mössbauer 8828 S L Bud'ko et al

measurements, Fe does not have moment in the structure and variation of the isomer shift and the quadrupole splitting with La concentration is consistent with variations of lattice parameters in the series.

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