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The Kondo contribution to the electrical resistivity in $UCu_{5-x}Ni_x$ and the non-Fermi liquid behaviour of UCu_4Ni

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Abstract

We report on electrical resistivity measurements performed on polycrystalline samples of $UCu_{5-x}Ni_x$ ($x = 0.25, 1$). In order to extract the Kondo contribution to the resistivity, the experiments were carried out over a wide temperature range (0.4–800 K). From the analysis of our results, we conclude that the Kondo temperature takes values of $T_K \sim 240$ K for $x = 1$ and $T_K \sim 245$ K for $x = 0.25$, and that for both Ni concentrations the dominant part of the remarkably high residual resistivity ($\rho(0) \sim 400\ \mu\Omega$ cm) corresponds to the Kondo contribution. These results are discussed in comparison with previous analysis of specific heat and magnetic susceptibility data that produced similar values of T_K. We interpret our results in terms of disorder-driven non-Fermi liquid behaviour for UCu_4Ni, as indicated by the anomalous temperature dependences of the electrical, thermal and magnetic properties previously observed in this compound.

1. Introduction

Amongst the unconventional low-temperature properties of the so-called 'non-Fermi liquids' (NFL) [1, 2], the electrical resistivity is possibly the most intriguing. In complete contradiction to Fermi liquid theory, which predicts a quadratic dependence of the electrical resistivity well below T_K, systems such as $Y_{1-x}U_xPd_3$ [1, 2], $CeCu_{5.9}Au_{0.1}$ [3], $UCu_{5-x}Pd_x$ [4], $U_{1-x}Th_xBe_{13}$ [5] and others display quasilinear ($\rho = \rho_0 \pm AT$) or fractional ($\rho = \rho_0 + AT^{1/2}$) temperature dependences of their low-temperature electrical resistivity. Another remarkable feature is their rather poor electrical conductivity. The high values of the electrical resistivities,

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usually well above 100 $\mu\Omega$ cm over the entire experimental temperature range, commonly found in such systems, in a conventional metal would indicate that the electronic mean free path is close to the interatomic distance. Thus, these materials can be included in the class of the 'bad metals': strongly correlated electron systems in the limit $k_F l = \mathrm{O}(1)$ (the Ioffe–Regel limit for the metallic state), whose reduced electrical conductivity prevents, in principle, the application of the Boltzmann theory of transport [6]. This class includes, apart from the Kondo alloys quoted above, others such as high-T_c superconductors, fullerenes, and ferromagnetic perovskites (e.g. $SrRuO_3$), all of them showing properties suggesting some sort of NFL behaviour [7].

Several models have been put forward in order to explain the NFL properties of these systems. As regards the Kondo alloys, we mention here the two-channel Kondo model, first proposed by Cox *et al* [8] for UBe_{13} and applied to $Y_{1-x}U_xPd_3$ by Seaman *et al* [1]. Other models consider fluctuations close to a quantum critical point at $T = 0$ K [2, 3, 9], or a disordered distribution of Kondo temperatures ('Kondo disorder') [10, 11]. Yet another model proposes the formation of magnetic clusters in the paramagnetic phase (the Griffiths phase) due to the competition between the Kondo effect and the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction in the presence of disorder [12]. In a previous study, we applied the Kondo disorder model to UCu_4Ni [13], a system that displays NFL properties [14]. This model has been successfully applied to UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ [11], which are isostructural to UCu_4Ni and the parent compound UCu_5. From fits of the magnetic susceptibility and the specific heat in terms of that model, we obtained an estimate of the Kondo temperature. In this paper, our aim is to obtain a value of the Kondo temperature independent of the particular model used to explain the nature of the NFL behaviour. To achieve this, we have analysed the high-temperature electrical resistivity in terms of a single-impurity Kondo effect, as is usual in Kondo lattices [15], and compared the results with those obtained using the Kondo disorder model.

2. Experimental details

Polycrystalline samples of $UCu_{5-x}Ni_x$ were prepared by arc melting stoichiometric amounts of the elements in an argon atmosphere, using high-purity starting materials. To improve sample homogeneity, the buttons were remelted and flipped over several times, and subsequently annealed in vacuum for one week at 900 °C. Powder x-ray diffraction, electron microscopy (SEM) and electron dispersive microanalysis (EDAX) experiments did not show evidence of any secondary phase. In the temperature range between 0.4 and 80 K, dc electrical resistivity measurements were performed in a top-loading ^{3}He Oxford Instruments cryostat using a four-probe method. Between 4.2 and 300 K, a conventional ^{4}He immersion cryostat and the same dc method were used. Finally, the high-temperature measurements (300–800 K) were performed in a vacuum furnace. The samples used in these experiments consisted of parallelepipeds with cross-sectional area of 1×1 mm^2 and length of approximately 12 mm, cut from the annealed polycrystalline ingots by spark erosion.

3. Results and discussion

In figures 1 and 2 we present the results of our electrical resistivity measurements for UCu_4Ni and $UCu_{4.75}Ni_{0.25}$. In both cases, the resistivity above 300 K can be fitted using the expression

$$\rho = \rho_0 + c_{ph}T - c\ln T. \tag{1}$$

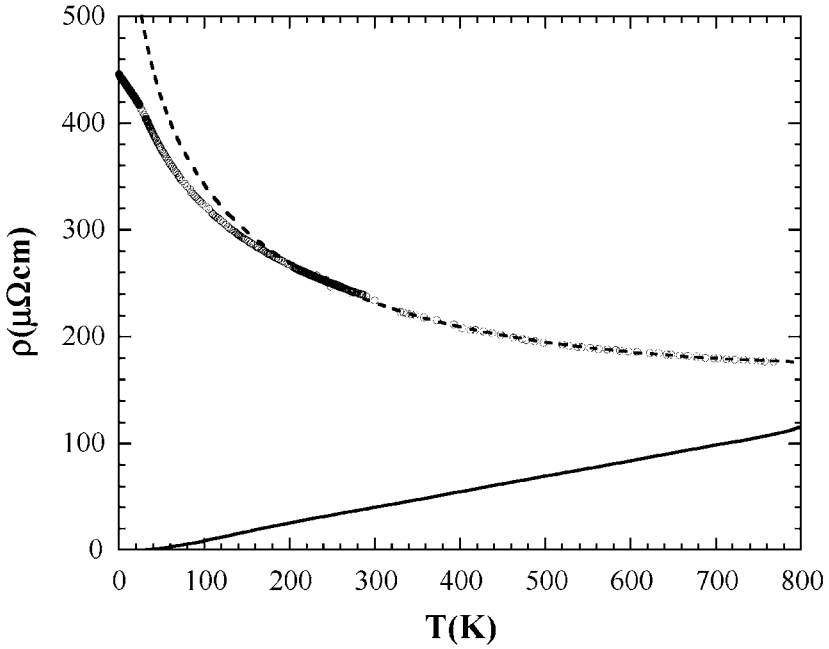

Figure 1. The temperature dependence of the electrical resistivity of UCu_4Ni measured over the temperature range between 0.4 and 800 K. The dashed curve is a fit to equation (1). The solid curve is the phonon contribution calculated using $\rho_{ph}(300\ \mathrm{K}) = 40\ \mu\Omega$ cm and a Debye temperature of 310 K.

In this temperature range ($T \geqslant \theta_D$), the linear term $c_{ph}T$ describes properly the contribution to the electrical resistivity due to electron–phonon scattering. The third term corresponds to the magnetic (Kondo) contribution. In both figures the solid curve represents fits using equation (1), with values $c_{ph} = 0.134(\pm 0.005)\ \mu\Omega$ cm K^{-1}, $\rho_0 = 905(\pm 12)\ \mu\Omega$ cm, $c = 125(\pm 3)\ \mu\Omega$ cm for UCu_4Ni and $c_{ph} = 0.176(\pm 0.009)\ \mu\Omega$ cm K^{-1}, $\rho_0 = 1144(\pm 24)\ \mu\Omega$ cm, $c = 167(\pm 5)\ \mu\Omega$ cm for $UCu_{4.75}Ni_{0.25}$.

Both fits were performed above 300 K. The goodness of such fits implies the relevance of single-impurity Kondo interactions in this range of temperatures for both systems. A single-impurity regime in the same temperature range has been also observed, and analysed in a similar way, for other classical Kondo lattice systems, such as URu_2Si_2 ($T_K = 370$ K) [15], for which comparable values of the fitting coefficients were obtained. In our case, the quality of the fit was not improved by adding a cubic correction to the simple linear term that accounts for the phonon contribution:

$$\rho_{ph}(T) = c_{ph}T(1 - BT^2). \tag{2}$$

Indeed, the introduction of such a term increases the errors associated with all the coefficients. The use of this correction, proposed by Mott and Jones [16], is appropriate if scattering processes on a narrow band close to the Fermi level, such as s–d processes in transition metals, make a significant contribution to the resistivity. Equation (2) has been used to describe deviations from the usual linear dependence in the high-temperature regime of certain rare-earth intermetallic compounds [17, 18]. The constant B related to this correction should be, in the simplest model, equal to $2/6T_F^2$ [16]. Our results point to a negligible contribution from this correction term for temperatures up to 800 K. This would be consistent with a

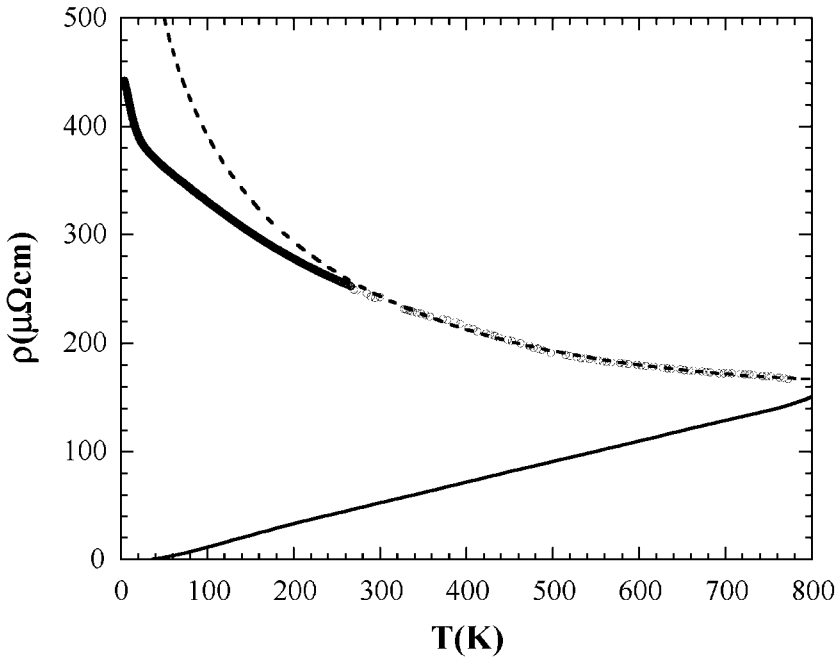

Figure 2. $\rho(T)$ versus T for $UCu_{4.75}Ni_{0.25}$ measured over the temperature range between 0.4 and 800 K. As in figure 1, the dashed curve is a fit to equation (1), and the solid line is the phonon contribution calculated using $\rho_{ph}(300\ \mathrm{K}) = 53\ \mu\Omega$ cm and $\theta_D = 310$ K.

characteristic Fermi temperature $T_F \geqslant 10\,000$ K, a value similar to that used by Bernal *et al* [11] and ourselves [13], to explain the NFL behaviour of UCu_4Pd, $UCu_{3.5}Pd_{1.5}$ and UCu_4Ni in terms of the Kondo disorder model.

In order to obtain a reliable estimate of the Kondo temperature, it is necessary to subtract the phonon contribution (ρ_{ph}) from the total electrical resistivity, over the whole temperature range of our measurements. We did not attempt to subtract the constant residual contribution due to structural defects, which in any case would not change the overall temperature dependence of the estimated magnetic contribution, and result in a small overestimate of the magnetic contribution to the resistivity. We estimate its value to be, like in other heavy-fermion systems, around $\rho_0 = 10\ \mu\Omega$ cm, i.e., about 2% of $\rho(0\ \mathrm{K})$. Above 300 K, the assumption of a linear temperature dependence of ρ_{ph}, as we have discussed above, is reasonable. However, the linear contribution obtained from the high-temperature fit does not give a proper description of $\rho_{ph}(T)$ in the low-temperature range of our measurements. Therefore, we have followed an alternative procedure to estimate $\rho_{ph}(T)$ over the whole experimental temperature range. First, we have used the coefficient c_{ph} to deduce the room temperature value of ρ_{ph} for both alloys. We obtain $\rho_{ph}(300\ \mathrm{K}) = 40\ \mu\Omega$ cm for UCu_4Ni and 53 $\mu\Omega$ cm for $UCu_{4.75}Ni_{0.25}$. These values can be compared to $\rho_{ph}(300\ \mathrm{K}) = 50\ \mu\Omega$ cm reported by van Daal *et al* for the isostructural non-magnetic compound UNi_5 [19]. With our values for $\rho_{ph}(300\ \mathrm{K})$, we then computed the phonon contribution to the electrical resistivity given by the Bloch–Grüneisen formula, using a value of 310 K for the Debye temperature θ_D. This value was deduced using the phonon term of the heat capacity reported in the literature for materials isostructural to UCu_4Ni ($ZrCu_5$ and UCu_4Pd; see [20]). Then, θ_D was scaled according to the molecular weights of UCu_4Ni and $UCu_{4.75}Ni_{0.25}$.

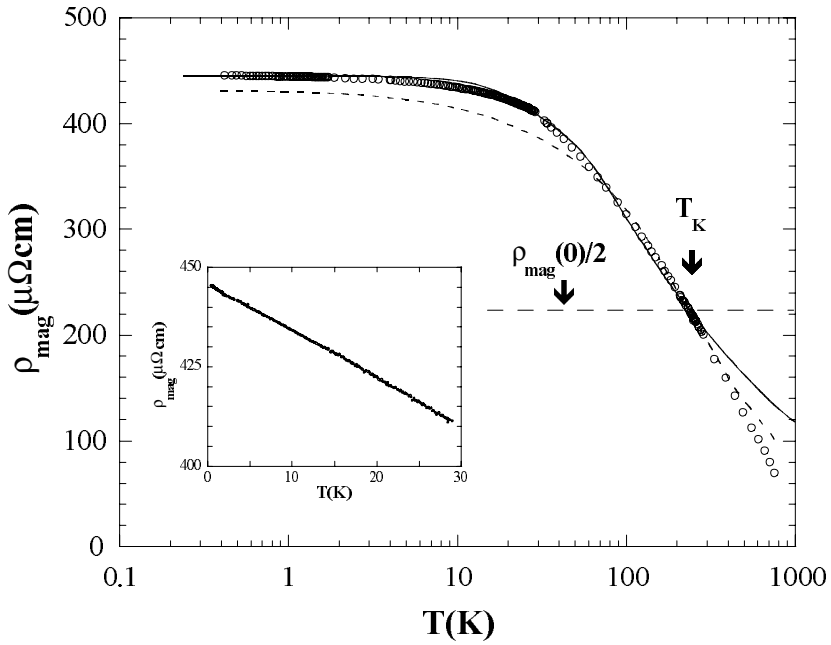

Figure 3. Magnetic contribution to the resistivity of UCu_4Ni plotted as ρ_{mag} versus log T, deduced by subtracting the calculated phonon term from the total electrical resistivity. The arrow indicates $T_K = 240$ K estimated using the criterion $\rho(T_K) = \rho_{mag}(0)/2$ (see the text). The dashed curve is the corresponding Hamann curve for $T_K = 240$ K and $s = 1/2$. The solid curve shows results of NRG calculations [24] for the same T_K. The inset displays the low-temperature magnetic resistivity, showing the linear temperature dependence characteristic of UCu_4Ni.

In figures 3 and 4 we display the magnetic (Kondo) contributions (ρ_{mag}) to the electrical resistivity after subtraction of the phonon backgrounds. The logarithmic temperature scale used in these figures reveals the $-\ln T$ dependence, indicative of Kondo interactions, which describes well the electrical resistivity over a broad temperature range between 200 and 800 K. Now we proceed to estimate the value of the Kondo temperature. For the conventional single-impurity Kondo effect, the usual way of determining T_K is from the 50% rise in the Kondo contribution to the resistivity ($\rho_{mag}(T_K) = \rho_{mag}(0)/2$) [21, 22]. From our results, we estimate $\rho_{mag}(0) = 445$ $\mu\Omega$ cm for UCu_4Ni and 435 $\mu\Omega$ cm for $UCu_{4.75}Ni_{0.25}$. We finally obtain $T_K \sim 240$ K for UCu_4Ni and 245 K for $UCu_{4.75}Ni_{0.25}$. As expected, the subtraction of the phonon contribution does not affect the linear temperature dependence observed in the low-temperature resistivity of UCu_4Ni [14] (see the inset to figure 3). To give further support to our discussion, we compare our result for the magnetic resistivity of UCu_4Ni with two theoretical curves calculated assuming $T_K = 240$ K. One of them is a calculation using the phenomenological expression due to Hamann for $s = 1/2$ [23], which describes quite well the results on diluted alloys of Fe and Cr in Cu, Au and CuAu alloys [21, 22], classical examples of Kondo systems. The other consists of exact numerical renormalization group (NRG) results [24] for the same T_K. Both curves describe well the overall temperature dependence of the experimental results, apart from the expected deviations at low and high temperatures. It is known that Hamann's curve is not valid in the low-temperature regime ($T \ll T_K$) [22, 23]. On the other hand, the results of NRG calculations [24] are in good agreement with our experimental results over the whole temperature range below T_K. The

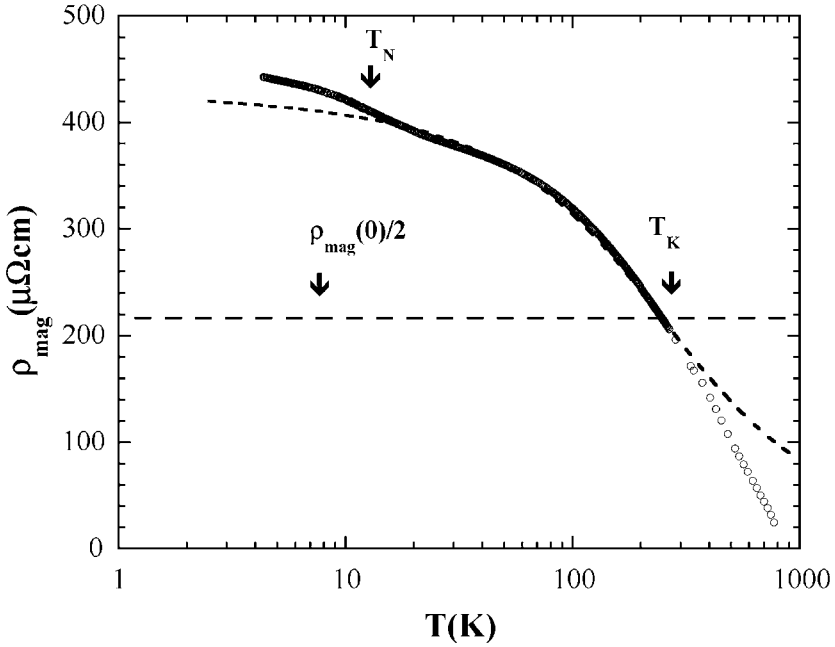

Figure 4. ρ_{mag} versus $\log T$ for $UCu_{4.75}Ni_{0.25}$, calculated as in figure 3. The arrows indicate $T_K = 245$ K estimated using the criterion $\rho(T_K) = \rho_{mag}(0)/2$, and the broad magnetic transition at $T_N \sim 12$ K. The dashed line is a Hamann curve for $T_K = 245$ K.

slight deviation from the experimental curve observed below $T_K/10$ is due to NFL effects on the electrical resistivity of UCu_4Ni. Results of NRG calculations have been reported to exhibit Fermi liquid behaviour at low temperatures [24]. At high temperatures we must take into the account the unavoidable uncertainty in the values of ρ_{mag} due to the subtraction of the calculated phonon term. The value of T_K obtained here from our electrical resistivity measurements is in good agreement with our own previous estimates obtained using the Kondo disorder model for UCu_4Ni: 390 and 260 K, obtained in each case from fits to the magnetic susceptibility and the heat capacity [13]. In our opinion, the Kondo disorder model applied to $UCu_{4-x}Ni_x$ provides a consistent explanation for the transport, thermal and magnetic properties of this system, considering a common T_K = O(300 K). On substitution of 20% Ni on copper sites, the magnetically ordered state that develops below $T_N = 16$ K in the parent compound UCu_5 fades away, and crystallographic disorder in the Cu sites (ligand disorder) induces local variations in the hybridization with the nearby U sites. This effect results in a spatial variation of the Kondo temperature [25], which is reflected in a distribution of T_K with an average value around 300 K, as estimated from specific heat and magnetic susceptibility measurements. This is in agreement with the clear single-ion Kondo behaviour observed in the high-temperature electrical resistivity, from which a similar value of the average T_K is obtained. On the other hand, the linear temperature dependence and the anomalously high values displayed by the low-temperature electrical resistivity simply reflect the destruction of the coherence of the Kondo lattice induced by disorder [26]. For $x = 0.25$, which above 20 K displays a single-ion Kondo behaviour very similar to that of UCu_4Ni, the amount of Ni impurity is not enough to destroy completely the magnetically ordered state, as the low-temperature upturn observed in figures 2 and 4 indicates. Antiferromagnetic order below $T_N = 12$ K for $x = 0.2$ has

been previously reported by other authors [27]. For $x = 0.25$, the absence of other features characteristic of the parent compound UCu_5, such as the maximum below T_N, followed by a rapid decrease as temperature is lowered [28, 29], are also consistent with the interpretation in terms of crystallographic disorder given above. In $UCu_{4.75}Ni_{0.25}$, the degree of crystallographic disorder is not enough to induce NFL behaviour in the electrical resistivity, which in any case would be masked by the broad magnetic transition taking place at $T_N \sim 12$ K. To reinforce our interpretation of the electrical properties of the $UCu_{5-x}Ni_x$ system, we point out that the compound UCu_4Ag, which develops antiferromagnetic order below $T_N = 20$ K, does not show any sign of NFL behaviour [28, 30], consistently with the arguments developed above. Whereas Cu and Ni have very similar ionic sizes, Ag is a much bigger ion. Considering that there are two different copper sites in the crystallographic structure of UCu_5, and that the minority sites (20%) have more free volume available, it is quite reasonable to expect a preferential occupation of these sites by Ag ions. Thus, compared to UCu_4Ni, UCu_4Ag should show a reduced degree of crystallographic disorder, as the sharpness of the antiferromagnetic transition observed at T_N clearly suggests. In consequence, as Kondo disorder is absent in this system, UCu_4Ag should not show NFL behaviour. Indeed, well below the magnetic transition the electrical resistivity of UCu_4Ag exhibits the quadratic temperature dependence expected for a conventional Fermi liquid [30].

4. Conclusions

In this work we have shown that the high-temperature electrical properties of UCu_4Ni and $UCu_{4.75}Ni_{0.25}$ can be explained in terms of single-impurity Kondo behaviour. After a proper subtraction of the phonon contribution to the electrical resistivity, we obtain T_K-values of 245 K for $UCu_{4.75}Ni_{0.25}$ and 240 K for UCu_4Ni. These values are in good agreement with those obtained applying the Kondo disorder model to our previous magnetic susceptibility and specific heat data for UCu_4Ni. All the electrical, thermal and magnetic properties of these materials are determined by a single energy scale $k_B T_K$. The unconventional NFL behaviour that develops at low temperature for UCu_4Ni can be explained in terms of crystallographic disorder that results in a distribution of Kondo temperatures about an average $T_K = \mathrm{O}(300\,\mathrm{K})$, equivalent to the effective single-ion Kondo temperature deduced from the electrical resistivity measurements presented in this work.

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References

[1] Seaman C L, Maple M B, Lee B W, Ghamaty S, Torikachvili M S, Kang J-S, Liu L Z, Allen J W and Cox D L 1991 *Phys. Rev. Lett.* **67** 2882
[2] Andraka B and Tsvelik A M 1991 *Phys. Rev. Lett.* **67** 2886
[3] von Löhneysen H, Pietrus T, Portisch G, Schlager H G, Schröder A, Sieck M and Trappmann T 1994 *Phys. Rev. Lett.* **72** 3262
[4] Andraka B and Stewart G R 1993 *Phys. Rev.* B **47** 3208
[5] Aliev F G, El Mfarrej H, Vieira S and Villar R 1994 *Solid State Commun.* **91** 775
[6] Emery V J and Kivelson S A 1995 *Phys. Rev. Lett.* **74** 3253
[7] Varma C M, Littlewood P B, Schmitt-Rink S, Abrahams E and Ruckestein A E 1989 *Phys. Rev. Lett.* **63** 1996

[8] Cox D L 1987 *Phys. Rev. Lett.* **59** 1240
[9] Tsvelik A M and Reizer M 1993 *Phys. Rev.* B **48** 9887
[10] Dobrosavljević V, Kirkpatrick T R and Kotliar G 1992 *Phys. Rev. Lett.* **69** 1113
[11] Bernal O O, MacLaughlin D E, Lukefahr H G and Andraka B 1995 *Phys. Rev. Lett.* **75** 2023
[12] Castro Neto A H, Castilla G and Jones B A 1998 *Phys. Rev. Lett.* **81** 3531
[13] López de la Torre M A, González J A, Izquierdo A, Vieira S, Ellerby M and McEwen K A 2000 *J. Appl. Phys.* **87** 5126
[14] López de la Torre M A, Ellerby M, Watmough M and McEwen K A 1998 *J. Magn. Magn. Mater.* **177–181** 445
[15] Schoenes J, Schönenberger C, Franse J J M and Menovsky A A 1987 *Phys. Rev.* B **35** 5375
[16] Mott N F and Jones H 1958 *The Theory of the Properties of Metals and Alloys* (London: Oxford University Press)
[17] Bauer E, Gratz E, Hauser R, Le Tuan, Galatanu A, Kottar A, Michor H, Perthold W, Hilscher G, Kagayama T, Oomi G, Ichimiya N and Endo S 1994 *Phys. Rev.* B **50** 9300
[18] Kaczorowski D, Andraka B, Pietri R, Cichorek T and Zaremba V I 2000 *Phys. Rev.* B **61** 15255
[19] van Daal D H, Buschow K H J, van Aken P B and van Maaren M H 1975 *Phys. Rev. Lett.* **34** 1457
[20] Andraka B 1994 *J. Alloys Compounds* **209** 43
[21] White R M and Geballe T H 1979 *Long Range Order in Solids* (New York: Academic)
[22] Loram J W, Whall T E and Ford P J 1970 *Phys. Rev.* B **2** 857
[23] Hamann D R 1967 *Phys. Rev.* **159** 570
[24] Costi T A, Hewson A C and Zlatić V 1994 *J. Phys.: Condens. Matter* **6** 2519
[25] Ohkawa F J 1990 *Phys. Rev. Lett.* **64** 2300
[26] Miranda E, Dobrosavljević V and Kotliar G 1997 *Phys. Rev. Lett.* **78** 290
[27] Chakravarthy R, Paranjpe S K and Madhav Rao L 1991 *Physica* B **174** 30–2
[28] Ott H R, Rudigier H, Felder E, Fisk Z and Battlogg B 1985 *Phys. Rev. Lett.* **65** 1595
[29] López de la Torre M A, McEwen K A, Ellerby M, Haworth C and Springford M 1995 *J. Phys.: Condens. Matter* **7** 9235
[30] Watmough M, McEwen K A, Ellerby M and López de la Torre M A 1998 *J. Magn. Magn. Mater.* **177–181** 469–71