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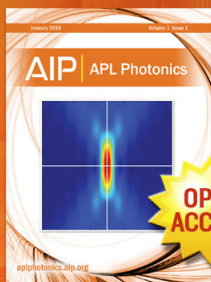Low Temp. Phys. **35**, 638 (2009); 10.1063/1.3224723

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Magnetization and specific heat in $U_{1-x}La_xGa_2$ and magnetocaloric effect in UGa_2

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We have investigated the properties of the ferromagnetic series $U_{1-x}La_xGa_2$. The magnetization results show a reduction of μ_{eff} and of T_c when x is increased. The electronic coefficient γ of the specific heat increases to a maximum of 260 mJ/Umol K^2 at $x=0.75$. This behavior is probably consequence of delocalization of $5f$ electrons, causing enhancement of the density of states. For $x=0.9$ the ordering disappears and a non-Fermi-liquid behavior is observed. UGa_2 also presented a significant magnetocaloric effect of $\Delta S_{mag}=-3.5$ J/kg K at 120 K and $H=7$ T which can be modified by chemical pressure. © *2005 American Institute of Physics.* [DOI: 10.1063/1.1854412]

UGa_2 crystallizes with the hexagonal AlB_2-type structure and order ferromagnetically below $T_c=125$ K.[1] Studies using both, single crystals and polycrystalline samples revealed that this compound is highly anisotropic and presents a high degree of localization of the $5f$ electrons, which is responsible for the magnetic ground state. Nevertheless, it has been observed that the replacement of Ga with other metals (Cu, Al) or semimetals (Ge) can significantly alter the ground-state properties.[2] When 25% of Ga is replaced with Cu, for example, this system presents spin-glass behavior[3,4] and when the Cu concentration reaches 50% (UGaCu), the compound order antiferromagnetically. These magnetic properties are related to the unit cell volume, which is significantly reduced with the increase of the Cu content. Therefore, controlled changes of the unit cell volume can induce modifications on the system ground state and the chemical substitution process in the U site can be a useful tool for this purpose, leaving the electronic character of the ligands unaltered. In this study we replaced U with La in order to check the changes induced in the UGa_2 properties. We report on magnetization and heat capacity measurements for the $U_{1-x}La_xGa_2$ series, with $0 \leq x \leq 1$. We also discuss trends on the magnetocaloric effect for UGa_2 and $UGa_{1.9}Al_{0.1}$.

The samples were prepared by melting the pure elements in an arc furnace in argon atmosphere and they where subsequently annealed at 700 °C for one week. X-ray diffraction analysis showed the formation of a solid solution for the whole series, with the unit cell volume increasing linearly with the La concentration.

The magnetization measurements were obtained with a MPMS system, in a temperature range between 1.7 K and 300 K. In Fig. 1 we show the magnetization for $U_{1-x}La_xGa_2$ with $x=0.1$, 0.25, 0.6, and 0.85, obtained with $H=500$ G, using a zero-field cooling (ZFC) and a field cooling (FC) sequence. The separation of the ZFC and FC curves are as-

sociated to domain wall changes, characteristic of anisotropic systems.[5]

The magnetization results for the series reveal that the temperature of the magnetic ordering is strongly reduced when x increased, as shown in the inset of Fig. 1. For concentrations above 90% of La the ordering disappears for $T > 2$ K. However, when substituting U for Y the ordering disappears already at 50% Y.[6] This strong reduction in T_c is a consequence of both, the dilution effect and of the weakening of the RKKY interaction due to the unit cell volume increase (negative pressure effect). However, the significant reduction of the effective moment from 3.4 μ_B for $x=0$ to 2.5 μ_B for $x=0.8$ shows that the volume expansion can be considered dominant.

In Fig. 2 we present the specific heat data for UGa_2 with $H=0$ and 7 T. The data were analyzed considering the elec-
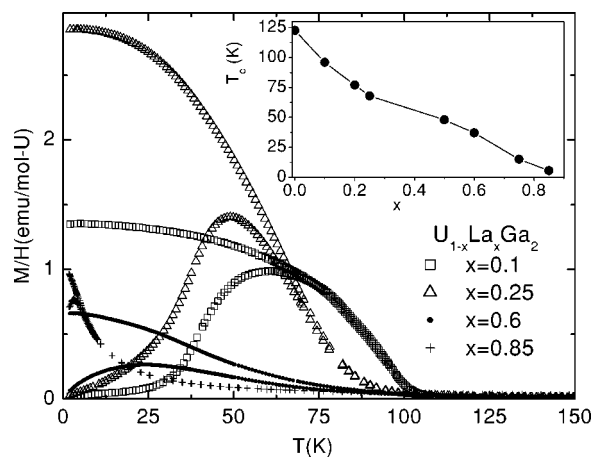

FIG. 1. ZFC and FC magnetization of $U_{1-x}La_xGa_2$ at $H=500$ G. The inset shows T_c as a function of the La concentration x, where the line is just a guide.
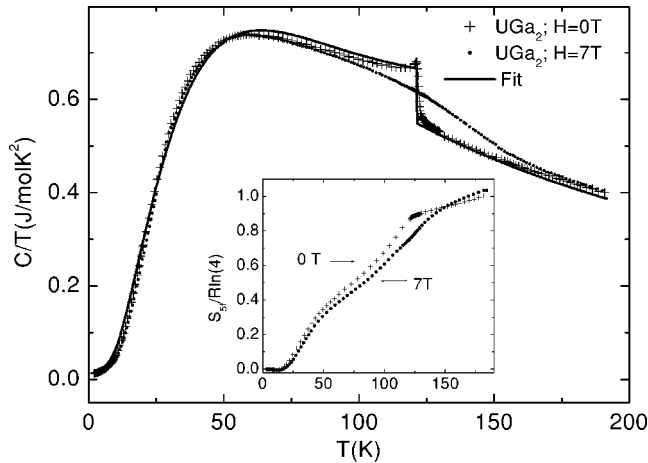
FIG. 2. C/T for UGa_2 in $H=0$ and 7 T. The solid line is the best fit to the contributions as explained in the text. The inset shows the magnetic entropy variation ΔS_{5f} for $H=0$ and 7 T.

FIG. 4. Magnetic specific heat (C/T) for $x=0.9$, showing the NFL behavior ($-\ln T$ dependence).

tronic, phonons, crystal field, and ferromagnetic ordering contributions. The solid line is the best fit with parameters: $\gamma=0.01$ J/K^2 mol, $\theta_D=230$ K, and two excited doublets for the crystal field ($\Delta_1=90$ K and $\Delta_2\sim500$ K). This result shows that the probable CF scheme is one of the possible solutions of the $U^{3+}(5f^3)$ configuration in hexagonal symmetry[7] where the two lower levels are doublets. A similar conclusion was reached by Radawansky and Kim-Ngam[8] using magnetic susceptibility data. In addition, the magnetic variation entropy reach a value around 90% of $R\ln 4$ at the transition (see inset of Fig. 2), that is very close to the expected value for a two doublet scheme [$S_{mag}=R\ln(2s+1)$, where s is the effective spin].

The remaining concentrations presented a Debye temperature varying between 215 K and 250 K but with the crystal field splitting essentially unchanged. On the other hand, the electronic coefficient increases with the La concentration reaching $\gamma\sim260$ mJ/K^2 molU at $x=0.75$ and reducing again subsequently, as shown in Fig. 3. The effective moment obtained with a modified Curie–Weiss law is also shown in this figure. For $x=0$, $\mu_{eff}\sim3.4$ μ_B [which is close

to the U^{3+} free ion ~3.62 μ_B (Ref. 8)] but it is reduced to 2.5 μ_B at $x=0.85$. This reduced μ_{eff} can either indicate a Kondo process or may be due to CF effect, but from our specific heat data analysis the CF is not changing with the concentration. Both, the enhanced γ and the reduced effective moment are indicative of a higher degree of hybridization of the $5f$ electron. In fact, for $x=0.9$ the FM transition is no longer observed and the magnetic specific heat present a $-\ln T$ dependence, as shown in Fig. 4. This specific heat dependence observed at low temperatures and in diluted Kondo systems is characteristic of non-Fermi-liquid (NFL) behavior. Similar results have been found for $(U,La)PdAl_3$.[9] In this latter compound the Neel temperature moved to zero for a large concentration of La with an increased unit cell volume, where a NFL behavior was found. We believe that the volume increase induced by magnetic dilution is responsible for the delocalization of the $5f$ electrons and weakening the long range ordering.

The proximity of the CF splitting with the magnetic transition, corresponding to a magnetic entropy of $R\ln 4$, brings to attention that this compound can be considered as a good candidate to present the magnetocaloric effect (MCE). This effect is promoted by the magnetic entropy variation when a magnetic field is applied. Usually significant MCE is achieved with fields above $2T$, which will affect the magnetic specific heat, with a consequent change in the magnetic entropy. For $x=0$ we obtained the magnetic entropy variation around T_c using $H=2$ and 7 T, as shown in Fig. 5. From this, we obtained a fair magnetocaloric effect of $\Delta S_{mag}=-3.5$ J/kg K. Although smaller than the giant effect observed at high temperatures in $Gd_5Si_2Ge_2$ (~-23 J/kg K at 280 K),[10] our result is comparable to the MCE found in UAs (Ref. 11) and UNi_2Si_2.[12] Also, recent results in MnAs show that pressure can be a very important parameter for the MCE.[13] For UGa_2, the chemical substitution can be an alternative way to alter the pressure with the corresponding response of the MCE. However, the magnetic dilution in $(U,La)Ga_2$, reduces the effective moment and causes a NFL behavior, which is not interesting for the MCE. Nevertheless, the chemical pressure effect on the MCE can be observed for $UGa_{1.9}Al_{0.1}$ for example, where an enhanced ΔS_{mag}
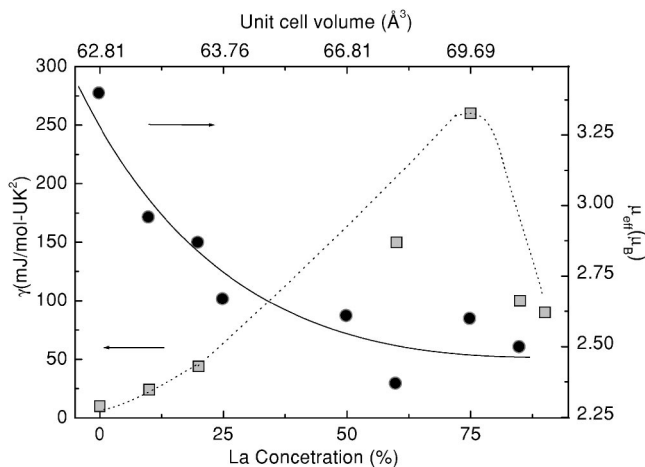

FIG. 3. The electronic coefficient γ and the effective moment μ_{eff} for the $U_{1-x}La_xGa_2$ series. The unit cell volume is shown at the top of the graph. The lines are just a guide to the eye.
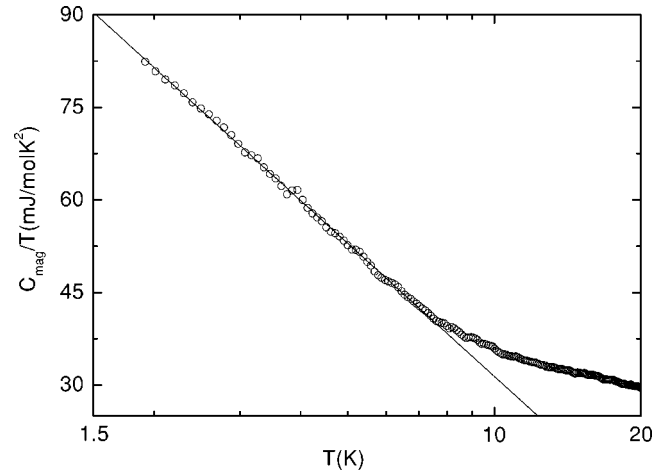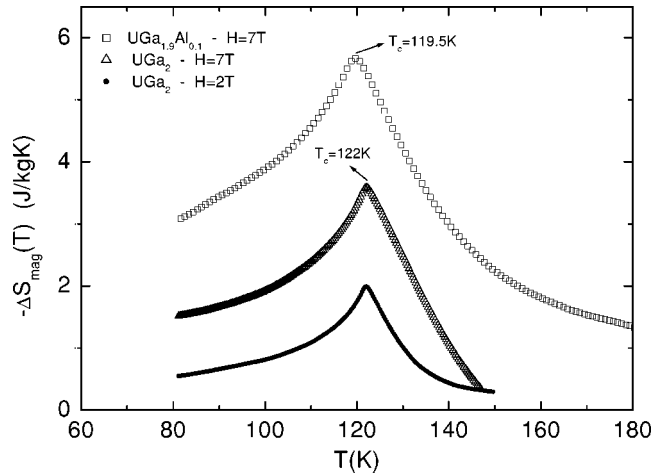
FIG. 5. Temperature dependence of $-\Delta S_{mag}$ for UGa_2 ($H=2$ T and 7 T) and $UGa_{1.9}Al_{0.1}$ ($H=7$ T).

$=-5.6$ J/kg K (see Fig. 4) justifies a further study of this material.

In conclusion, the increase of the unit cell volume induced by substitution of U for La causes a weakening of the long range ordering, with a subsequent appearing of a NLF behavior. The results also show that UGa_2 is an interesting material to study the MCE.

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