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Evidence for fluctuating Fe-moments in RFe_2Ge_2 ($R = Lu, Yb$)

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Abstract

We have reinvestigated the magnetic properties of $LuFe_2Ge_2$ by means of susceptibility $\chi(T)$ and specific heat $C(T)$ measurements on flux-grown single crystals. The observation of a well-defined Curie–Weiss behavior in $\chi(T)$ above 80 K indicates the presence of fluctuating Fe-moments with $\mu_{eff} = 2.9\mu_B$, as already proposed to be the case in $YbFe_2Ge_2$. A magnetic Fe character is supported by LDA calculations. Anomalies in $\chi(T)$ and $C(T)$ confirm the presence of a phase transition at $T_0 = 9$ K, which we suggest to be a spin density wave.

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It is presently generally believed that in the compounds RT_2X_2 (R: rare earth, T: transition metal, X: Si, Ge) the 3d-elements Fe, Co and Ni are nonmagnetic [1]. For T = Fe very early experiments claimed a spontaneous magnetization of the Fe-sublattice, but later experiments gave conclusive evidence for a non-magnetic Fe-state in those compounds which were investigated more carefully. However, investigations of the germanides are rather scarce. Very recently, we started an investigation of $YbFe_2Ge_2$, and found to our surprise a much larger value of the effective moment at high temperature than expected for Yb^{3+}, which pointed to a fluctuating Fe-moment of the order of $3.0\mu_B$ [2]. During the course of our investigation, Avila et al. [3] reported an extensive investigation of the series RFe_2Ge_2 for all rare earth elements with the exception of La, Ce, Pm, Eu and Yb. While they notice an enhanced Sommerfeld coefficient and an enhanced temperature-independent susceptibility in YFe_2Ge_2 and $LuFe_2Ge_2$, they apparently did not consider the possibility of fluctuating Fe-moments. We therefore decided to perform our own investigation of $LuFe_2Ge_2$ in order to have a reliable basis for the analysis and interpretation of our $YbFe_2Ge_2$ results.

The susceptibility $\chi(T)$ and the specific heat $C(T)$ were measured on flux-grown $LuFe_2Ge_2$ single crystals using a MPMS and a PPMS, respectively. Our results are very similar to those of Ref. [3]. As pointed out in Ref. [3], ferromagnetic Fe-based impurity phases can be a significant problem in these RFe_2Ge_2 compounds. Extensive measurements at different magnetic fields indicated that in our samples the ferromagnetic contribution was rather small, with a saturated moment of the order of $3 \times 10^{-4}\mu_B$ per formula unit (~100 ppm Fe). At $B = 5$ T the error in the susceptibility induced by this contribution is less than 1% and thus does not affect our analysis. In Fig. 1, we show $\chi(T)$ at $B = 5$ T applied along the two crystallographic directions. Along both directions $\chi(T)$ increases with decreasing temperatures below 400 K, evidences a weak maximum around $T \approx 50$ K and shows at 9 K a small but well-defined anomaly. The susceptibility is slightly anisotropic, being 20% larger along the basal plane than along the c direction.

In order to gain more insight into the T-dependence of $\chi(T)$, we plot $1/\chi$ versus T (inset of Fig. 1). This plot reveals a very well defined Curie–Weiss law, with almost the same slope for both field directions. A fit through the data between 80 and 400 K results in an effective moment

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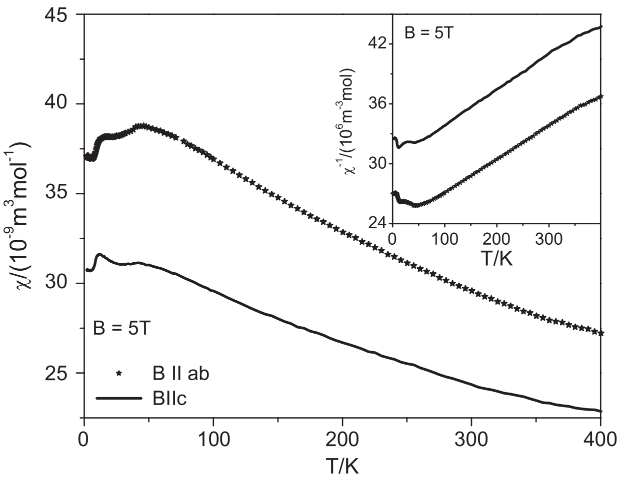

Fig. 1. Temperature dependence of the susceptibility and (inset) inverse susceptibility of $LuFe_2Ge_2$ single crystals.

of $4.15(\pm 0.2)\mu_B$ per formula unit, which corresponds to an effective moment of $2.9\mu_B$ per Fe, almost the same value as our estimation in $YbFe_2Ge_2$. The Curie–Weiss temperatures are rather large, $\Theta \approx 800\,\mathrm{K}$. Thus our results and our analysis confirm the presence of a fluctuating paramagnetic Fe-moment in the RFe_2Ge_2 compounds. Avila et al. already recognized in $LuFe_2Ge_2$ the presence of a phase transition at $T_0 = 9\,\mathrm{K}$ evidenced by anomalies in $\chi(T)$ and $C(T)$, but they left its nature open. In order to gain a deeper insight into this phase transition, we looked in more details in $\chi(T)$ and $C(T)$ at low T. In Fig. 2, we compare $C(T)$ and $\chi(T)$ data at low field (after subtraction of the T-independent ferromagnetic contribution) along both directions. These data already evidence a rather strong decrease of $\chi(T)$ at T_0 for the field along the basal plane, while for $B\|$c the anomaly looks much weaker. Even after the subtraction of an impurity Curie contribution, determined from a $\chi(T)\cdot T$ versus T plot for $T < 5\,\mathrm{K}$, the anomaly is still much more pronounced for $B\|$ab than for $B\|$c. Thus our analysis indicates that the transition at T_0 is connected with an anisotropic decrease of the susceptibility, of the order of 15% for $B\|$ab and only 3% for $B\|$c. This anisotropy, which is also visible in the data of [3], suggests that the phase transition is antiferromagnetic.

The corresponding anomaly in the specific heat is rather small but well visible. The Sommerfeld coefficient estimated from an extrapolation of $C(T)/T$ to $T = 0\,\mathrm{K}$ amounts to $\gamma = 74\,\mathrm{mJ/K^2mol}$. Using this γ value and an average low temperature susceptibility $\chi = 35\times 10^{-9}\,\mathrm{m^3/mol}$, we get a Wilson ratio $R = K\cdot\chi_0/\gamma = 2.7$ ($K = \pi^2 k_B^2/3\mu_0\mu_B^2$), which is only slightly enhanced. This (as well as the large Θ value) indicates that $LuFe_2Ge_2$ is not close to a ferromagnetic instability, but is consistent with the proximity of an antiferromagnetic instability. Thus our results evidence in $LuFe_2Ge_2$ a Curie–Weiss behavior of the susceptibility, which indicates the presence of fluctuating Fe moments with $\mu_{\mathrm{eff}} = 2.9\mu_B$ at high temperatures, as already proposed for $YbFe_2Ge_2$. Preliminary LDA calculations confirm that in the RFe_2Ge_2 compounds Fe is at the border of being magnetic. The calculated ground state energy is slightly lower for a configuration with a small ferromagnetic Fe-moment than for the paramagnetic state.

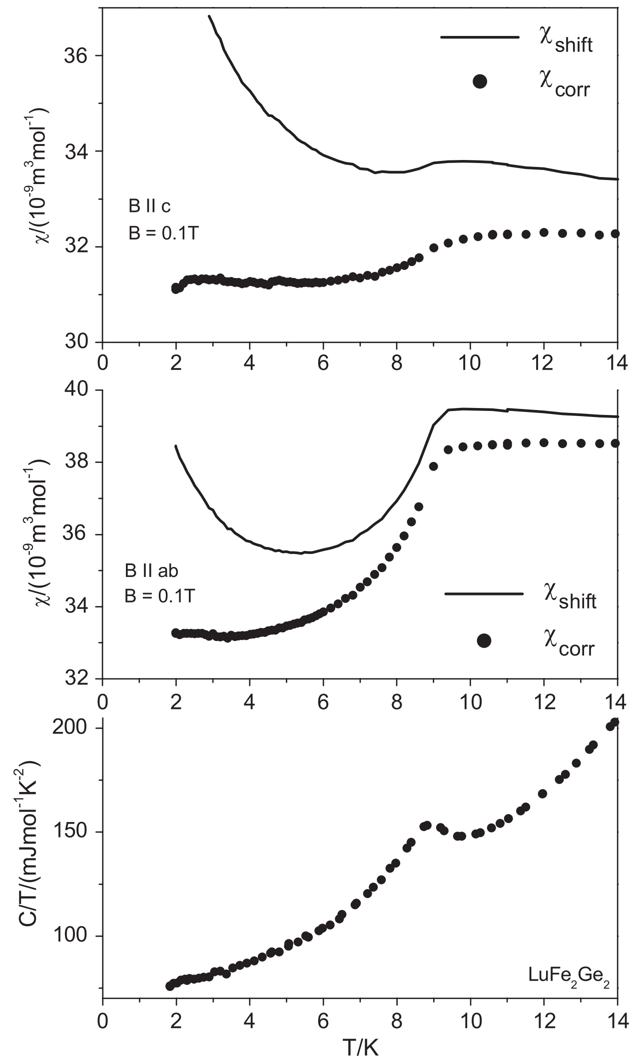

Fig. 2. Anomalies in $C(T)$ and $\chi(T)$ at the phase transition at 9 K. χ_{shift} was obtained by subtracting a T-independent ferromagnetic contribution from the measured data. For χ_{corr} we further subtracted the low-T Curie contribution (see text).

We further confirm the presence of a phase transition at $T_0 = 9\,\mathrm{K}$ in $LuFe_2Ge_2$. The strong anisotropy of the related anomaly in $\chi(T)$ as well as the observation of a fluctuating Fe-moment at higher temperatures and results from LDA calculations suggest this phase transition to be magnetic ordering of Fe, likely a spin density wave.

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