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PRESSURE EFFECT ON MAGNETIC PROPERTIES OF UGa_3

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The magnetic susceptibility, χ, of the itinerant antiferromagnetic compound UGa_3 was studied under pressure up to 2 kbar in the temperature range 64–300 K. The measured pressure derivative of the Néel temperature is found to be $dT_N/dP = -1.1$ K/kbar. In order to analyze the experimental magnetovolume effect values, $d\ln\chi/d\ln V$, the volume dependent electronic structure of UGa_3 has been calculated *ab initio* in a paramagnetic phase by employing a relativistic full-potential linear muffin tin orbital method and including an external magnetic field self-consistently. The calculations revealed a predominance of itinerant uranium f-states at the Fermi energy, as well as a large orbital contribution to χ.

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Itinerant $5f$-electron magnetism in UGa_3 has been suggested on the basis of the temperature dependence of its magnetic susceptibility (a rise in $\chi(T)$ near $T_N = 67$ K and a lack of Curie–Weiss behavior in the temperature range up to 900 K [1–3]). This suggestion is also supported by the results of a scalar-relativistic band-structure calculation [4]. In this contribution further evidence for the itinerant electron nature of the magnetic properties of UGa_3 is obtained from experimental magnetic susceptibility studies under pressure and *ab initio* relativistic calculations of the volume dependent field-induced magnetization. The role of orbital magnetism and its pressure dependence is discussed.

The magnetic susceptibility of UGa_3 was studied under helium gas pressure up to $P = 2$ kbar at 77.5 and 300 K using the Faraday method [5]. The $\chi(P)$ dependences appeared to be linear, and the resulting values of $d\ln\chi/dP$ are -6.8 ± 0.5 and -5.4 ± 0.5 Mbar^{-1} at T = 77.5 K and 300 K, respectively. The corresponding volume derivatives, $d\ln\chi/d\ln V$, evaluated from $d\ln\chi/dP$,

TABLE

Magnetic susceptibility χ (in 10^{-3} emu/mole) and its volume derivative in UGa_3.

	Experiment	Theory	
	$T = 77.5$ K	orbital	spin
χ	1.95	4.3	−2.4
$d\ln\chi/d\ln V$	4.5 ± 0.5	4.4	1.7

are listed in Table. In addition, the $\chi(T)$ dependence for UGa_3 was measured in the vicinity of T_N for two different pressures, yielding the pressure derivative $dT_N/dP = -1.1 \pm 0.2$ K/kbar. The corresponding value $d\ln T_N/d\ln V = 11 \pm 2$ is comparable to the analogous derivative value, 26, found for itinerant antiferromagnetic chromium [6]. Also, a clearly visible peak has been observed in $\chi(T)$ at 10 K, which can be attributed to another antiferromagnetic phase transition.

In order to analyze the observed $d\ln\chi/d\ln V$ values, the electronic structure of the paramagnetic phase of UGa_3 was calculated *ab initio* by employing the LMTO-ASA [7] and the full-potential LMTO (FP-LMTO) [8] methods. In the latter method the charge density and potential were allowed to have any shape inside the muffin-tins as well as in the interstitial region. The calculations are all electron, fully relativistic and include the spin–orbit and Zeeman operators in each variational step, in line with Ref. [9]. The orbital polarization correction [10] was also included. The exchange and correlation potential was treated in the local spin density approximation using the von Barth–Hedin parametrization [11]. The density of states, $N(E)$, and other parameters, including the Stoner multi-band exchange integral, I [10], were calculated for a number of lattice parameters close to the experimental one ($a = 4.248$ Å). The main contributions to the total density of states at the Fermi level, E_F, come from the U $5f$-states ($\approx 70\%$) and the Ga $4p$-states ($\approx 20\%$), which overlap in energy. This hybridization is sufficiently strong to form a band of itinerant uranium f-states in the energy range ≈ 0.2 Ry. The values of $N(E_F, T = 77\ \mathrm{K}) \equiv N$ and $d\ln N/d\ln V$ were found to be 91 Ry^{-1} and 1.9, respectively. (For finite temperatures the effect of "smearing" through the Fermi–Dirac distribution function has been taken into account.) The calculated linear coefficient of the specific heat, $\gamma = 2\pi^2 {k_B}^2 N(E_F)/3$, is in agreement with the experimental value $\gamma_{exp} = 52$ mJ/(mole K^2) [1], providing many-body enhancement factor, λ, is about 2.

In order to estimate the spin magnetic susceptibility, the Stoner model has been employed, in which electron–electron interactions manifest themselves in magnetic properties through the enhancement of the Pauli spin susceptibility. In our fully relativistic calculations for the experimental lattice parameter the Stoner criterion was not quite fulfilled within both LMTO-ASA and FP-LMTO methods, whereas according to the scalar-relativistic calculation [4] $IN(E_F)$ appeared to be ≈ 1.3, due to a higher value of $N(E_F)$. Nevertheless, the calculated high value of the Stoner enhancement factor, $S \approx 10$, is apparently related to the antiferromagnetic transitions observed in UGa_3, and multiplied with the $N(E_F)$ yields a

spin susceptibility close to the experimental data on $\chi(T)$, extrapolated to zero temperature: $\chi(0) \approx 2 \times 10^{-3}$ emu/mole. On the other hand, it can be expected that in the actinides, where the spin–orbit energy dominates over the Zeeman spin polarization energy, the orbital magnetic moment is larger than, and anti-parallel to, the spin moment (see e.g. [9, 12]). Indeed, as can be seen in Table it actually takes place in UGa_3, where the spin moment is anti-parallel to the applied field. The presented contributions to the magnetic susceptibility were derived from the corresponding magnetic moments, calculated in an external field of 10 T. The orbital susceptibility of the U site appeared to be almost twice the value of the spin susceptibility, and the resulting total susceptibility matches favourably the experimental value. Remarkably, with the orbital polarization correction omitted our calculations yield very close values for anti-parallel orbital and spin moments, which result in a too small total susceptibility.

The preliminary result of the calculated atomic volume effect on χ in UGa_3 is also given in Table. It appeared to be more pronounced for the orbital contribution to χ, presumably due to the quenching of the induced orbital moment with increasing width of the f-band. The agreement with the experimental data looks promising, but more detailed calculations are necessary due to close proximity of the induced spin-polarized state to the spontaneous magnetic ordering. Actually, for lattice parameters $a \geq 4.24$ Å the calculated magnetic moment is rising gradually to a large value ($\approx 1\mu_B$), close to the measured value of the total moment in the antiferromagnetic state ($0.8\mu_B$ [2]). This is however not an induced magnetization since it turns out that a ferromagnetic state with a such moment appeared to be more stable for higher a values, than the considered above paramagnetic state.

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