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Effect of pressure on magnetic susceptibility of $UCu_{5-x}Au_x$ alloys

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Abstract

The magnetic susceptibility of UCu_3Au_2 and UCu_3Au_2 has been measured at 78 and 290 K under hydrostatic pressures up to 2 kbar. The pressure dependence of the susceptibility is correlated with the change in the paramagnetic Curie temperature. For both compounds the $d\Theta/dP$ value was determined to be about -0.2 ± 0.05 K kbar^{-1}.

Keywords: Magnetic susceptibility; Hydrostatic pressure; RKKY-type interaction

1. Introduction

The study of magnetic and transport properties of the $UCu_{5-x}Au_x$ system, crystallizing in the cubic $AuBe_5$-type structure [1], shows that doping of the initial antiferromagnetic UCu_5 ($T_N = 15$ K) causes an appreciable change in the magnetic ordering temperature. The substitution of one Au atom for one Cu atom nearly doubles the Néel temperature. With further growth of the Au content, T_N decreases and finally the antiferromagnetic state disappears at $x \geqslant 0.3$. These observed properties have been explained within the framework of a competition between the Kondo effect and RKKY exchange interactions [2]. At high enough temperatures the magnetic susceptibility χ of $UCu_{5-x}Au_x$ alloys shows the modified Curie–Weiss behaviour

$$\chi(T) = \chi_0 + \frac{C}{T-\theta} \quad (1)$$

It appears that the effective magnetic moment per U atom stays roughly constant ($\mu_{eff} \approx 3\ \mu_B$) and the paramagnetic Curie temperature Θ (about -100 K for UCu_5) increases monotonically (decreases in modulus) with increasing Au content. In this system the strength of hybridization between f and ligand sp electrons, which influences the magnetic behaviour, is believed to be correlated with the preferential occupation of Au atoms at the 4*c* sites of the $AuBe_5$-type structure [2]. The substitution of Au for Cu increases appreciably the crystal lattice parameter *a* and hence also the spacing between the U–U and U–Au(Cu) atoms. The increase in the interatomic distances also modifies the effective exchange interactions between the 5f electrons [1].

In this paper the effect of hydrostatic pressure on the magnetic susceptibility of some alloys of the $UCu_{5-x}Au_x$ system will be presented.

2. Experimental

Polycrystalline samples of UCu_3Au_2 and UCu_2Au_3 (about 0.5 g) were spark cut from the ingot used earlier [1]. Their magnetic susceptibility was measured under helium gas pressures up to 2 kbar at 290 and 78 K using a pendulum magnetometer placed directly in the pressure cell [3]. The relative measurement error was no larger than $\pm0.05\%$. The experimental dependence $\chi(P)$ appears to be linear for these two compounds. As an example, in Fig. 1 we display such a dependence for UCu_2Au_3. The resulting derivatives $d\ln\chi/dP$ of the compounds are given in Table 1.

3. Discussion of results

According to Eq. (1), the pressure effect can be described as

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Table 1
Magnetic parameters and their pressure derivatives for UCu_3Au_2 and UCu_2Au_3

	UCu_3Au_2	UCu_2Au_3
$d(\ln \chi)/dP$ ($Mbar^{-1}$)		
($T=78$ K)	-1.3 ± 0.3	-1.4 ± 0.3
($T=290$ K)	–	-0.7 ± 0.3
χ (10^{-3} e.m.u. mol^{-1})		
($T=78$ K)	6.16	7.00
($T=290$ K)	–	3.24
$d(\ln C)/dP$ ($Mbar^{-1}$)	–	0.0 ± 0.5
$d\Theta/dP$ (K $kbar^{-1}$)	-0.21 ± 0.05	-0.20 ± 0.05

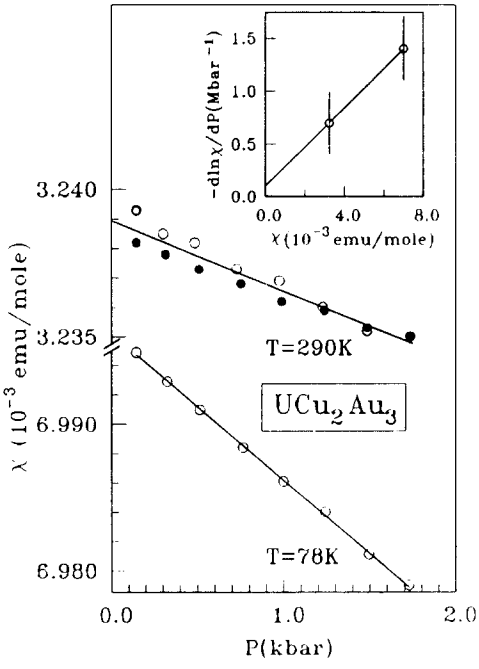

Fig. 1. Pressure dependence of the magnetic susceptibility for UCu_2Au_3 at 78 and 290 K. The closed and open circles indicate data of two different measurements. The inset shows the derivative $d(\ln \chi)/dP$ as a function of the susceptibility.

$$\frac{d(\ln \chi)}{dP} \approx \frac{d(\ln C)}{dP} + \frac{\chi}{C}\frac{d\Theta}{dP} \quad (2)$$

if the contribution χ_0 and its dependence on pressure are disregarded (the possible χ_0 value for the alloys considered here does not exceed 10% of χ at $T \leqslant 300$ K). As seen in Eq. (2), the $d(\ln \chi)/dP$ value for different temperatures is a linear function of χ, and $d\theta/dP$ and $d(\ln C)/dP$ can be obtained from the corresponding parameters of the $d(\ln \chi)/dP$ vs. χ straight line. The experimental values of $d(\ln \chi)/dP$ for UCu_2Au_3 at 78 and 290 K, which are plotted against χ in the inset of Fig. 1, suggest that the effect of pressure on the paramagnetic Curie constant is small ($d(\ln C)/dP = 0\pm0.5$ M bar^{-1}). This result is indirectly supported by the constancy of the uranium effective moment in the $UCu_{5-x}Au_x$ system. Thus the effect of pressure on the magnetic susceptibility in the system investigated is mainly due to the change in the paramagnetic Curie temperature under pressure:

$$\frac{d\Theta}{dP} \approx \frac{C}{\chi}\frac{d(\ln \chi)}{dP} \quad (3)$$

The resulting values of $d\Theta/dP$ practically coincide for the two compounds (see Table 1). They were estimated using experimental values of the susceptibility and an adopted value of $C \approx 1.2$ K e.m.u. mol^{-1} (corresponding to $\mu_{eff}=3\ \mu_B$ per U atom [1]).

The obtained value of $d\Theta/dP \approx -0.2\pm0.05$ K $kbar^{-1}$ may be used to estimate the effect of the change in the lattice parameter a on the Θ value in $UCu_{5-x}Au_x$ due to the substitution of Au for Cu atoms, i.e. due to the chemical pressure of Au atoms. According to Ref. [1], $d(\ln a)/dx \approx 0.02$ and is equivalent to the pressure change $dP/dx = -3Bd(\ln a)/dx \approx -75$ kbar/x when the average component bulk modulus [4] over the formula unit is taken as the estimate of its value for the compound: $B \approx 1.25$ Mbar. Then the expected change in Θ on doping should be $d\Theta/dx = 15\pm5$ K/x.

The real change in Θ with increasing Au content in $UCu_{5-x}Au_x$ alloys can be estimated by comparison of the inverse susceptibilities for different x values in Eq. (1) at constant temperature:

$$\frac{\Delta\Theta}{\Delta x} \approx \frac{\Delta(1/\chi)}{\Delta x} \quad (4)$$

The χ_0 variations in the alloys are neglected because they are small (less than 0.8×10^{-3} e.m.u. mol^{-1}) in comparison with the total suceptibilities. Averaging the $1/\chi$ data [1] at 100 and 300 K over $x=0$, 1, 2 and 3 gives $d\Theta/dx = 16\pm5$ K/x. This value is close to that estimated from the effect of the change in the lattice parameter on Θ. This permits us to conclude that the variation in the lattice parameter in $UCu_{5-x}Au_x$ alloys is the predominant mechanism causing the variation in the magnetic characteristics, because the latter are mainly dependent on the interatomic spacing. It should be noted that this conclusion applies to the magnetic properties of the system at relatively high temperatures ($T \geqslant 78$ K) where the RKKY-type mechanism of interactions between the 5f moments of uranium predominates. However, at low temperatures the interactions are complicated by a manifestation of another mechanism, e.g. a Kondo effect, which is suggested by the various dependences of T_N and Θ on the composition [1,2]. To separate the predominant interaction at low temperatures and to elucidate the role of the atomic distance variation, systematic studies of $T_N(P,x)$ are

necessary, but are at present confined to $T_N(P)$ data only for UCu_5 [5].

In conclusion, we note that in $UCu_{5-x}Au_x$ the average value of the derivative $d(\ln \Theta)/dV = -4 \pm 1.5$ appears to be close to that for UCd_{11} (-4.2 ± 1). The latter value follows from measurements of the pressure effect on the magnetic susceptibility [6] when the bulk modulus of pure Cd ($B \approx 0.5$ Mbar [4]) is taken as that of UCd_{11}. Such a value seems to be typical for dilute uranium systems with the predominating RKKY-type antiferromagnetic interaction.

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