Magnetic properties and phase transitions of RAlxGa2-x (R = Tb, Ho)

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Colloque C5, *supplément au n°* 5, *Tome* 40, *Mai* 1979, *page* C5-159

Magnetic properties and phase transitions of RAl_xGa_{2-x} (R = Tb, Ho)

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Résumé. — On a étudié la structure cristalline et des propriétés magnétiques des composés RAl_xGa_{2-x} (R = Tb, Ho : $0 \leqslant x \leqslant 2{,}0$) par diffraction des rayons X et des neutrons, microanalyse et mesures de la susceptibilité.

Abstract. — Crystal structure and magnetic properties of RAl_xGa_{2-x} (R = Tb, Ho ; $0 \leqslant x \leqslant 2.0$) were investigated by means of X-ray and neutron diffraction, microprobe analysis and measurements of magnetic susceptibility.

Introduction. — RAl_2 crystallize with cubic structures of $MgCu_2$ type (Laves phase) and order predominantly ferromagnetic [1]. On the other hand the antiferromagnetic RGa_2 occur with hexagonal AlB_2 type structure. Many other compounds of Al and Ga are isostructural [2]. Magnetic structures of RGa_2 were investigated at Grenoble [3]. We determined [4] the influence of Ga substitution by Al in RGa_2 on crystal structures and magnetic properties. One anticipates enhanced exchange associated with modified interatomic distances and band structures. Similar investigations concerning RAlGa (R = Tb, Ho) were published recently by Gignoux and Asmat [5]. In contrast to this study our results furnish clear evidence for magnetic phase transitions in RAlGa and substantially increased Néel temperature of TbAlGa compared with $TbGa_2$. Coexistence of two helical phases is found in $TbAl_{0.5}Ga_{1.5}$.

1. Sample preparation, metallographic and X-ray investigations.

1. Sample preparation, metallographic and X-ray investigations. — The samples were synthesized in an arc furnace in purest argon atmosphere from 99.999 % gallium (Alusuisse), 99.99 % aluminium (Alusuisse), 99.9 % holmium (Johnson, Matthey and Co., Ltd.) and 99.9 % terbium (Johnson, Matthey and Co., Ltd.) and remelted several times. The ingots were enclosed under vacuum in tantalum containers, homogenised at 1 000 °C for 24 h and finally annealed at 600 °C for 72 h. The samples were examined metallographically, by microprobe analysis and X-ray diffraction. The specimens prove to be homogeneous and correspond well to the nominal compositions.

Two phases exist in the composition range RAl_xGa_{2-x}. The AlB_2 phase exists over a range of $x = 0.0$ to 1.6, the $MgCu_2$ phase from $x = 1.25$ to 2.0, i.e. both phases coexist in the range $x = 1.25$ to 1.6. Concerning substitution of Ga by Al the cell constants of the AlB_2 phase show a remarkably nonlinear dependence, whereas the change of the $MgCu_2$ cell constants is small and linear (cf. figure 1).

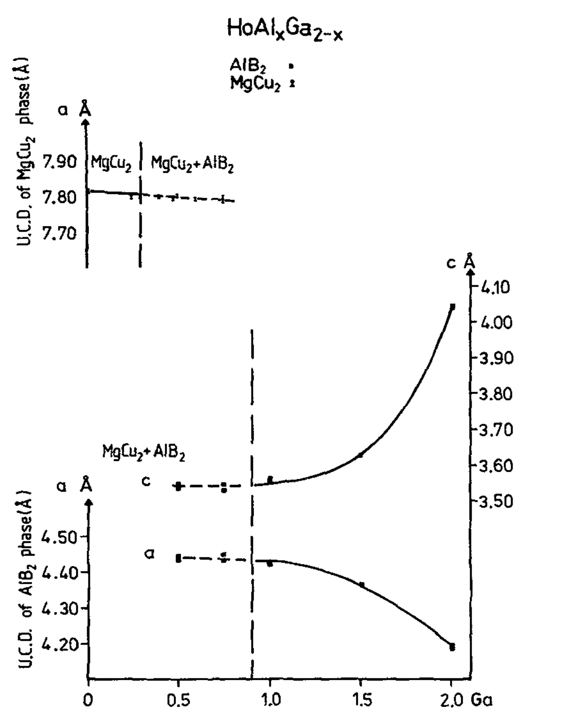


Fig. 1. — Cell constants (Å) *versus* Ga-content in $HoAl_xGa_{2-x}$ series.

2. **Magnetic susceptibility.** — Figure 2 shows an example of the susceptibility measurements of TbAlGa, where (similar to HoAlGa) two transitions can be recognised.

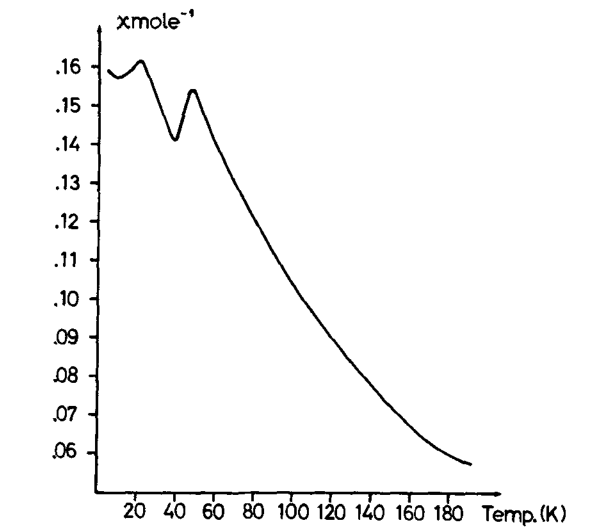
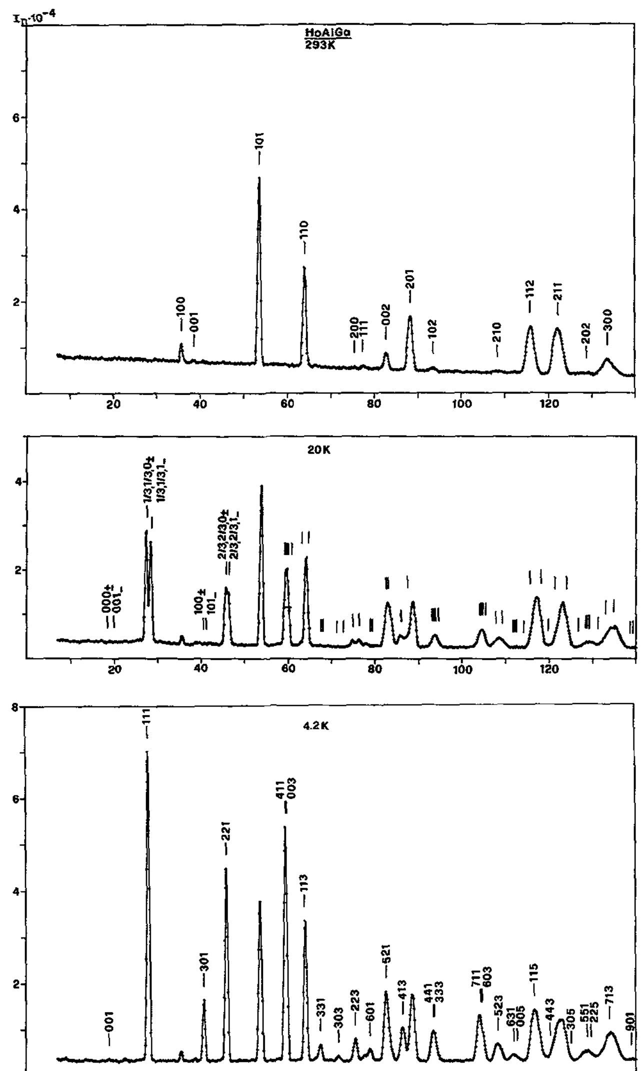


Fig. 2. — Susceptibility measurements of TbAlGa compound (emu).



3. **Neutron diffraction studies.** — Powder samples of RAl_xGa_{2-x} (cf. table I) were investigated by neutron diffraction at Würenlingen. Illustrative, absorption corrected patterns (wavelength 2.35 Å) are shown in figure 3. The nuclear intensities confirm the AlB_2-structure model with statistical distribution of Al and Ga on B sites. RAl_xGa_{2-x} (R = Tb, Ho: $x \leqslant 1$) order antiferromagnetically (cf. figure 3 and table I). At $x = 1$ Néel temperatures are considerably larger compared to $x = 0$. The low temperature magnetic structure of HoAlGa (cf. figure 4) corresponds to Shubnikov group P_c6/mcc and is similar to $CsCoCl_3$ [6]. The incommensurate configurations are spiral structures of triangular type within (001) planes [phase angles $\pm$ (120°, 240°)], with propagation direction **c** (**K** = [1/3, 1/3, q]) and q increasing systematically with composition x.

Table I. — *Neutron results for* RAl_xGa_{2-x}. T_N = *Néel température,* T_t = *temperature of magnetic phase transition,* MS = *magnetic structure.* Δ *denotes triangular moment configuration with undetermined orientation within* (001) *planes.* μ = *ordered magnetic moment. Standard deviations within parentheses refer to the last digit.* * *more stable above* 15 K.

R		Tb		Ho
x	0	0.5	1.0	1.0
T_N [K]	20.0 [3]	19.3 [5]	47.0 [5]	30.0 [3]
T_t			23.0 [5]	17.8 [2]
q	0.178	{0.339 0.419*	0.5 [< T_t] 0.483 [> T_t]	0.5 [< T_t] 0.482 [> T_t]
MS	Δ	Δ	Δ [> T_t] Δ, + − − [< T_t]	Δ [> T_t] + − − [< T_t]
μ_x [μ_B]			2.6 [1] [4.4 K]	0 [4.2 K]
μ_z			6.2 [1]	8.2 [1]
μ			6.7 [1]	8.2 [1]

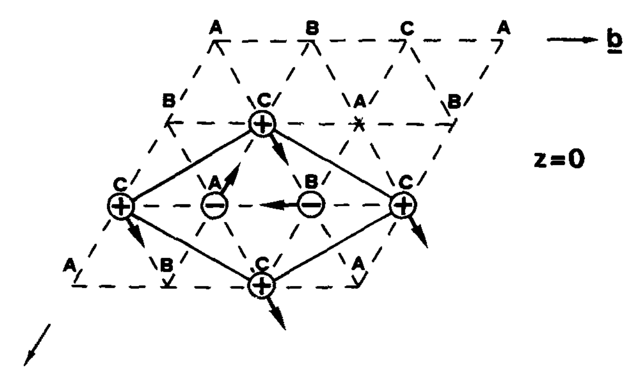


Fig. 4. — Low temperature magnetic structure of RAlGa (R = Tb, Ho) [moments reversed at $z = 1/2$]. Signs indicate z components.

Fig. 3. — Neutron diffraction patterns of HoAlGa. At 4.2 K magnetic reflections are indexed with respect to the magnetic ($a_m = 3\,a$, $c_m = 2\,c$) unit cell. $0_\pm = \pm\, q$.

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