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MAGNETIC ORDER OF THE COMPOUND SERIES

RE₆(MₙFe₁₋ₓ)₃₃ (RE = Y, Gd)

H. R. KIRCHMAYR (*) and W. STEINER (**)

Résumé. — La magnétisation et la susceptibilité des composés isostрукturels (structure Th₆Mn₂₃) de la composi-
tion RE₆(MₙFe₁₋ₓ)₃₃ (RE = Y, Gd) ont été déterminées de 4 °K à 1 200 °K. Les températures de Curie Tc de RE₆Mn₂₃ et
RE₆Fe₂₃ varient entre 468 et 486 °K. Dans le cas de Y₆(MₙFe₁₋ₓ)₃₃ pour x = 0.5 à 0.7 et à une température de 4 °K
un ordre magnétique a pu être observé ; si Y est remplacé par Gd, Tc = 120 °K. Les courbes μ(x) où le nombre de magné-
tons de Bohr, μ, est déduit de l’aimantation) montrent des minima profonds. Les résultats peuvent être expliqué en suppo-
sant un ordre antiparallèle entre les moments Mn et Fe et un ordre parallèle des moments Gd aux moments Mn.

Abstract. — The magnetization and the susceptibility of isostructural compounds (Th₆Mn₂₃-structure) of the composi-
tion RE₆(MₙFe₁₋ₓ)₃₃ (RE = Y, Gd) have been determined from 4 °K to 1 200 °K. The Curie-temperatures Tc of the
RE₆Mn₂₃ and the RE₆Fe₂₃ compounds (RE = Y, Gd) are in the range from 468 °K to 486 °K. In the case of Y₆(MₙFe₁₋ₓ)₃₃
at 4 °K for x = 0.5-0.7 no magnetic order at all is observed ; if Y is replaced by Gd, Tc = 120 °K. Also the μ(x) curves
show deep minima (the Bohr-magneton number μ is calculated from the magnetization). The results can be explained
by assuming an antiparallel ordering of the Mn- and Fe-moments to each other and a parallel ordering of the Gd-moments
to the Mn-moments.

I. Introduction. — In intermetallic compounds of rare earth metals (RE) with 3d-transition metals [1], the magnetic interaction between the 4f- and 3d-moments can be studied in detail [2]. The samples have been prepared by high-frequency melting without any annealing procedure and were checked by X-ray analysis [3]. All RE₆(MₙFe₁₋ₓ)₃₃ samples have the Th₆Mn₂₃ structure with a smooth change of only 3 % of the lattice spacings with x [1]. A statistical distribution of the Mn and Fe atoms must be assumed.

II. Magnetic measurements. — The susceptibility and the magnetization were determined at temperatures from 4 °K to 300 °K in fields up to 10 kOe by a self-compensating pendulum balance [4]. Above 300 °K another Faraday-balance was used.

From the magnetization at different fields H the saturation magnetization σ(0, ∞) at 0 °K and H = ∞ was extrapolated. From this value the Bohr-magneton number μ was calculated for the formula unit RE₆(MₙFe₁₋ₓ)₃₃/6. The Curie temperature Tc was determined by extrapolating the σ²(H) curves to the σ²(H = 0) curves which then were plotted versus T and extrapolated to σ²(H = 0) = 0.

III. Results and discussion. — Figure 1 shows that in the system Y₆(MₙFe₁₋ₓ)₃₃ the Curie temperatures for x = 0 and x = 1, i. e. for Y₆Fe₂₃ and Y₆Mn₂₃ are nearly the same, namely approx. 484 °K and 486 °K resp. For x = 0.48 to 0.68 no magnetic ordering (determined by magnetization measurements) is present at 4 °K. The statistical replacement of the Fe-atoms in Y₆Fe₂₃ by Mn-atoms or of the Mn-atoms in Y₆Mn₂₃ by Fe-atoms lowers the Curie temperature in a drastic way. The same behavior is found for Gd₆(MₙFe₁₋ₓ)₃₃, however, the lower limit for Tc is 120 °K at x = 0.5 (Fig. 2). The presence of the Gd-moments therefore increases the stability of the magnetic order.

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In the case of the Y₆(MₙFe₁₋ₓ)₃₃ compounds the concentration dependence of the magnetic moment μ resembles the Tc-dependence. The magnetic moment changes in a non-linear way with x. Also for the series Gd₆(MₙFe₁₋ₓ)₃₃, a non-linear change of μ with x is observed. The minimum of μ is shifted to x = 0.1, while the minimum of Tc is found to occur at approximately x = 0.5. The absolute value of μ for Y₆(MₙFe₁₋ₓ)₃₃ is larger on the Fe-side, while for Gd₆(MₙFe₁₋ₓ)₃₃ the contrary is true. The susceptibility is for all x-values temperature dependent and allows the calculation of effective values.

By assuming as a first approximation localized magnetic moments, the following model can be used to explain the results (Fig. 3). For the RE₆Fe₂₃ and RE₆Mn₂₃ compounds it can be concluded from measurements of the magnetization and of the susceptibility.

Fig. 1. — Curie-temperature Tc and magnetic moment μ of Y₆(MₙFe₁₋ₓ)₃₃, (**) after [5].
as a function of temperature that a parallel, ferromagnetic coupling of the 3 d-moments is present [2]. In the RE₆Mn₂₃ compounds the RE-moments couple mostly antiparallel to the Mn-moments. The compound Gd₆Mn₂₃ seems to be an exception [6].

By Mössbauer-measurements on Y₆Fe₂₃ the hyperfine field on the different lattice points of the Fe-atoms could be measured [2, 5]. It was found that neither the hyperfine field was the same at the 4 different lattice sites, occupied by Fe-atoms, nor was the electron-density at the Fe-nuclei the same. It was also shown by neutron-diffraction on Y₆Mn₂₃ [7] and Ho₆Fe₂₃ [8] that the crystallographically non-equivalent 3 d-atoms are non-equivalent with respect to the direction and magnitude of their magnetic moments. Nevertheless the temperature dependence of the reciprocal susceptibility of Y₆Mn₂₃ and Y₆Fe₂₃ is typical ferromagnetic.

If one assumes for simplicity that all Fe-atoms in Y₆Fe₂₃ and Gd₆Fe₂₃ and all Mn-atoms in Y₆Mn₂₃ and Gd₆Mn₂₃ have the same magnetic moment, one can construct figure 3. The total magnetic moment of YFe₂₃/₆ (μ = 7.43 μ₀) and YMn₂₃/₆ (μ = 2.09 μ₀) is then distributed uniformly on the different atoms located at the lattice sites no. 1, 2, 3 and 4. Since the number of atoms present at these lattice sites is 32, 32, 24 and 4, all atoms of one of these sites must contribute in the same ratio to the total moment. The basic assumption of the proposed model is now that the Mn-atoms couple their moment antiparallel to the resulting total Fe-moment. Physically important is only the absolute value of the magnetic moment.

If now instead of Y-atoms Gd-atoms are present, these atoms contribute a well-localized and therefore constant magnetic moment to the total magnetic moment independent of the x-values. It is an experimental fact that the μ-value for Y₆Fe₂₃ is larger than for Gd₆Fe₂₃ (see Fig. 1 and 2). Therefore the Gd-moments must couple antiparallel to the Fe-moments. If we now assume that the contribution of the Fe- and Mn-atoms to the total moment as a function of x is the same in Gd₆(Mn, Fe)₂₃ as in Y₆(Mn, Fe)₂₃, then the broken line represents the magnetic moment of Gd₆(Mn, Fe)₂₃ as a function of x. Again only the absolute value is important.

This general scheme of coupling of the magnetic moments, as given in Figure 3, can in fact explain essential experimental results. The magnetic moment of Y₆Fe₂₃ is larger than that of Gd₆Fe₂₃, however, the magnetic moment of Gd₆Mn₂₃ is larger than that of Y₆Mn₂₃. The compensation point is for Y₆(Mn, Fe₁₋ₓ)₂₃ near x = 0.7, for Gd₆(Mn, Fe₁₋ₓ)₂₃ near x = 0.1. The shortcomings of this model are obvious. The assumption of the same magnetic moment of all Fe-atoms resp. Mn-atoms at different lattice sites is not correct, as has been discussed above. A detailed analysis of the value of the moments at different lattice sites can, however, only be based e.g. on more detailed neutron-diffraction investigations. These moments are also obviously x-dependent and therefore the μ(x) curves are not linear. Also the fact that there exists a non-vanishing value of μ for Gd₆(Mn, Fe₁₋ₓ)₂₃ for all x-values cannot be explained by the simple model. The general agreement between the model
and the experimental results, however, is so good that the basic assumptions seem to be valid. These are an antiparallel ordering of the Mn- and the Fe-moments to each other and a parallel ordering of the Gd-moments to the Mn-moments. The details of the coupling of the different magnetic moments are now evaluated by other experiments, including neutron diffraction, Mössbauer-effect and NMR.

References