

# Nature of the magnetism of the $d$ subsystem in $\text{RMn}_2$ compounds

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An investigation was made of the magnetic properties and thermal expansion of cubic Laves phases with the formula  $(\text{R}, \text{Y})\text{Mn}_2$ , where  $\text{R} = \text{Gd}, \text{La}, \text{or Lu}$ . At low temperatures the stable phase of yttrium-rich compositions was tetragonally distorted and had the antiferromagnetic structure. The cubic phase of  $(\text{Gd}, \text{Y})\text{Mn}_2$  and  $(\text{Y}, \text{Lu})\text{Mn}_2$  systems was stable with 40 and 5 at. % of gadolinium and lutetium, respectively. The magnetic ordering of the manganese subsystem in cubic  $\text{RMn}_2$  compounds was found to be the result of the  $f$ - $d$  exchange interaction and was interpreted using a model of band metamagnetism of the  $d$  electrons. A neutron diffraction study of  $\text{Y}_{0.9}\text{Lu}_{0.1}\text{Mn}_2$  (with zero  $f$ - $d$  exchange) demonstrated the absence of magnetic ordering in the manganese subsystem.

## INTRODUCTION

Laves phases with the formula  $\text{RMn}_2$  have two subsystems, the rare-earth (with localized magnetic moments) and manganese (band) subsystems. In compounds containing heavier earths (which will be the subject of the present paper) the rare-earth subsystem is always magnetically ordered at low temperatures, whereas the behavior of the manganese subsystem depends on the atomic number of the rare earth: it is magnetically ordered if  $\text{R} = \text{Gd}, \text{Tb}$ , but paramagnetic if  $\text{R} = \text{Dy}, \text{Ho}, \text{Er}, \text{Tm}$  (Refs. 1–3).

This behavior of the manganese subsystem is not yet fully understood and there are two alternative views on this subject. According to one of them,<sup>4,5</sup> magnetism of the manganese subsystem is primarily due to the intrinsic  $d$ - $d$  exchange interaction and appears if the distance between neighboring manganese atoms exceeds a certain critical value  $r_c$  (this distance corresponds to the lattice parameter  $a_c \approx 7.54 \text{ \AA}$ ). The origin of the critical value of the Mn–Mn distance is not discussed in Refs. 4 and 5. The treatments given in Refs. 4 and 5 ignore also the influence of the rare-earth subsystem on the state of the  $d$  electrons in  $\text{RMn}_2$ . In particular, if we use the critical distance model, we find that it is difficult to explain the disappearance of the magnetic order in the manganese subsystem observed in mixed  $\text{Tb}_{1-x}\text{Y}_x\text{Mn}_2$  compounds in the range  $0.14 \leq x \leq 0.9$  (Ref. 6) when the crystal lattice parameters are greater than the critical value  $a_c$ . Moreover, this model fails to answer the question why ordering of the manganese subsystem in  $\text{GdMn}_2$  and  $\text{TbMn}_2$  is a first-order phase transition.

The second model postulates that the intrasublattice  $d$ - $d$  interaction is insufficient for spontaneous splitting of the  $d$  band of manganese and that the magnetic order observed in this subsystem is induced by the magnetization created by the rare earth.<sup>1,6</sup> It is postulated that the manganese subsystem has the properties of a band metamagnet (like the cobalt subsystem in  $\text{RCO}_2$  Laves phases<sup>7</sup>): the magnetic order appears abruptly when a certain critical field  $H_c$  is reached.

The effective field acting on the manganese  $d$  subsystem in  $\text{RMn}_2$  depends on the  $g$  factor  $g_R$  and on the total angular momentum  $J_R$  of the rare earth<sup>1</sup>:

$$H_{\text{R-Mn}} \propto (g_R - 1)J_R. \quad (1)$$

It exceeds  $H_c \sim 5 \times 10^6 \text{ Oe}$  in the case of  $\text{TbMn}_2$  and  $\text{GdMn}_2$

and is less than  $H_c$  for intermetallics containing other rare earths. Therefore, the manganese subsystem is ordered only in terbium and gadolinium compounds (Ref. 1).<sup>11</sup>

In this model the abrupt magnetic disordering of the manganese subsystem as the temperature of  $\text{GdMn}_2$  or  $\text{TbMn}_2$  increases can readily be explained by a reduction in  $H_{\text{R-Mn}}$  (because the momentum of the rare-earth subsystem decreases). We can also account for the paramagnetism of the manganese subsystem in  $\text{TbMn}_2$  when terbium is replaced partly with yttrium (this happens because of a reduction of  $H_{\text{R-Mn}}$  in mixed compositions to values less than  $H_c$ ).

Since a reduction in the atomic number of a heavy rare earth reduces  $H_{\text{R-Mn}}$  (because of reduction in  $J_R$ ) and also the lattice parameter (because of the lanthanoid compression), both these models yield an approximately the same dependence of the magnetic moment of manganese in pure  $\text{RMn}_2$  compounds on the atomic number of the rare earth (Fig. 1).

Experimental data on  $\text{YMn}_2$  are regarded by the authors of the first model as important supporting evidence. In spite of the absence of the magnetic moment in the case of yttrium ( $H_{\text{R-Mn}} = 0$ ), this compound is antiferromagnetic at temperatures below 90 K and the transition to the antiferromagnetic state is of the first order, exactly as in  $\text{RMn}_2$  ( $\text{R} = \text{Gd}$  and  $\text{Tb}$ ).<sup>8</sup> However, we would like to draw attention to the fact that the antiferromagnetic transition in  $\text{YMn}_2$  differs in many respects from the magnetic transitions in  $\text{GdMn}_2$  and  $\text{TbMn}_2$  where the manganese subsystem is magnetically ordered.

Firstly, in the case of  $\text{YMn}_2$  this transition is characterized by a considerably larger (by a factor of 4) magnetic volume anomaly  $\omega_s = \Delta V/V$ , although the magnetic moment of manganese in all three compounds is approximately the same (according to the band model,<sup>9</sup> we have

$$\omega_s = k\mu_{\text{Mn}}^2, \quad (2)$$

where the coefficient of proportionality  $k$  represents the elastic and magnetoelastic properties of the crystal lattice).

Secondly,  $\text{YMn}_2$  exhibits a wide ( $\sim 100 \text{ K}$ ) range of coexistence of the antiferromagnetic (low-temperature) and paramagnetic (high-temperature) phases, whereas in the case of  $\text{GdMn}_2$  and  $\text{TbMn}_2$  the hysteresis of the magnetic transition is considerably less.<sup>1,10</sup>

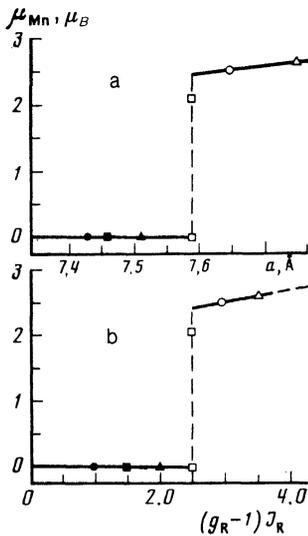


FIG. 1. Magnetic moment of the manganese subsystem in  $\text{RMn}_2$  compounds with  $\text{R} = \text{Tb}$  ( $\circ$ ),  $\text{Gd}$  ( $\Delta$ ),  $\text{Dy}$  ( $\square$ ),  $\text{Ho}$  ( $\blacktriangle$ ),  $\text{Er}$  ( $\blacksquare$ ), and  $\text{Tm}$  ( $\bullet$ ) deduced from NMR data<sup>4</sup> plotted as a function of the lattice parameter (a) and of  $(g_R - 1)J_R \propto H_{\text{R-Mn}}$  (b).

Thirdly, the magnetic ordering of manganese in  $\text{YMn}_2$  is accompanied by giant (for magnetic phase transitions) tetragonal distortions of the cubic lattice:  $(a - c)/a > 2 \times 10^{-3}$  (Ref. 10). In the case of  $\text{GdMn}_2$ , where gadolinium is in the  $S$  state and makes no contribution to the spontaneous anisotropic magnetostriction, the magnetoelastic distortions are less than  $5 \times 10^{-5}$  (Ref. 1).

All this raises doubts about the magnetic nature of the phase transition in  $\text{YMn}_2$  and it seems to us more appropriate to consider the suggestion made in Ref. 10 that in the case of  $\text{YMn}_2$  we are dealing with a structural transition and the magnetic ordering is a consequence of such a transition.

This account demonstrates the need for experiments which would exclude the possibility of interpretation of the results by both models: the model of the critical distance and the band metamagnetism model. With this in mind, we investigated the system  $\text{Gd}_{1-x}\text{Y}_x\text{Mn}_2$  in which the terminal compositions are characterized by a magnetically ordered manganese subsystem (lattice parameters  $a > a_c$ ) and the  $f$ - $d$  exchange interaction decreases on increase in  $x$ . The final selection of one or the other model is also affected fundamentally by the proof of existence or absence of mixed  $(\text{R}', \text{R}'')\text{Mn}_2$  compounds which exhibit zero  $f$ - $d$  exchange interaction when their lattice parameter is greater than the critical value  $a_c$  and which are paramagnetic at low temperatures. We shall search for these compounds in the  $\text{Y}_{1-x}\text{Lu}_x\text{Mn}_2$  and  $\text{Y}_{1-x}\text{La}_x\text{Mn}_2$  systems.

### SAMPLES AND MEASUREMENT METHODS

We used polycrystalline samples synthesized as described earlier.<sup>11</sup> The susceptibility and the crystal lattice parameters were determined in the temperature range 5–300 K by methods described in Ref. 1. We used a diffractometer with a multidetector recording system mounted in horizontal channels in a nuclear reactor. The neutron wavelength was 2.424 or 1.513 Å. In our calculations we used the following amplitudes of the nuclear neutron scattering:

$$b_{\text{Y}} = 0.76 \cdot 10^{-4} \text{ \AA}, \quad b_{\text{Lu}} = 0.73 \cdot 10^{-4} \text{ \AA}, \quad b_{\text{Mn}} = -0.37 \cdot 10^{-4} \text{ \AA}.$$

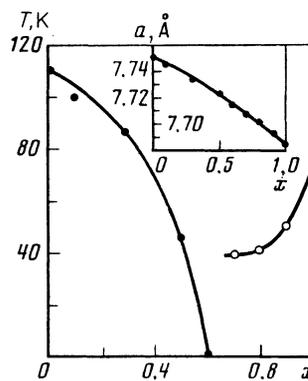


FIG. 2. Composition dependence of the temperature of appearance of the volume anomaly in  $\text{Gd}_{1-x}\text{Y}_x\text{Mn}_2$  compounds: ( $\circ$ ) transition accompanied by tetragonal distortions; ( $\bullet$ ) low-temperature cubic phase. The inset shows the dependence of the lattice parameter on the yttrium concentration at  $T = 295 \text{ K}$ .

### EXPERIMENTAL RESULTS

#### a) System with a magnetic rare earth: $\text{Gd}_{1-x}\text{Y}_x\text{Mn}_2$

As pointed out already, the manganese subsystem of  $\text{Tb}_{1-x}\text{Y}_x\text{Mn}_2$  is paramagnetic in a wide range of compositions ( $0.14 \leq x \leq 0.9$ ), although the interatomic Mn–Mn distances in these compositions are greater than the critical value.<sup>6</sup>

The  $\text{Gd}_{1-x}\text{Y}_x\text{Mn}_2$  system behaves in a qualitatively similar manner. Figure 2 shows the composition dependence of the magnetic ordering temperature of the manganese subsystem of these compounds, determined from the x-ray diffraction data on the appearance of the volume anomaly. In the range  $x \geq 0.7$  this transition is accompanied by tetragonal distortions of the crystal lattice and compositions with high gadolinium concentrations ( $x < 0.6$ ) have cubic structure below and above the transition.

It is clear from Fig. 2 that the replacement of gadolinium with yttrium in  $\text{GdMn}_2$  strongly reduces the ordering temperature of the manganese subsystem so that in  $\text{Gd}_{0.4}\text{Y}_{0.6}\text{Mn}_2$  it is at least lower than 5 K (which was the lowest temperature at which measurements were carried out). Clearly, in compositions close to  $x = 0.6$  the manganese subsystem is disordered although for these compositions the lattice parameter  $a = 7.715 \text{ \AA}$  is considerably greater than  $a_c = 7.54 \text{ \AA}$ .<sup>2)</sup>

Therefore, the results obtained for the  $\text{Tb}_{1-x}\text{Y}_x\text{Mn}_2$  and  $\text{Gd}_{1-x}\text{Y}_x\text{Mn}_2$  systems demonstrate that the concept of the critical parameter  $r_{\text{Mn-Mn}}$  is inappropriate in the case of mixed  $\text{RMn}_2$  compounds with magnetic rare earths and indicate the importance of the  $f$ - $d$  exchange interaction in the magnetic properties of the manganese subsystem. However, our experiments failed to reveal completely the role of the  $d$ - $d$  exchange interaction. In any case, in addition to the band metamagnetism, the absence of magnetic order in the manganese subsystem in mixed  $\text{R}_{1-x}\text{Y}_x\text{Mn}_2$  compounds can also be due to a competition between the  $f$ - $d$  and  $d$ - $d$  exchange interactions, so that the total effective field acting on the manganese subsystem vanishes (exchange-compensated paramagnetism<sup>13</sup>).

We must draw attention to the fact that in the case of yttrium-rich compositions ( $x \geq 0.7$ ) the volume anomaly is accompanied by tetragonal distortions of the crystal lattice

(exactly as in pure  $Y\text{Mn}_2$ ), whereas in compounds with higher gadolinium concentrations ( $x < 0.6$ ) the crystal structure remains cubic also below the transition (exactly as in pure  $\text{GdMn}_2$ ). Moreover, the volume anomalies and regions of coexistence of the high- and low-temperature phases are different for these two groups of compounds. A similar situation occurs also in mixed  $\text{Tb}_{1-x}\text{Y}_x\text{Mn}_2$  compounds.<sup>6</sup>

We can assume that there are two types of phase transition in  $\text{R}_{1-x}\text{Y}_x\text{Mn}_2$  ( $\text{R} = \text{Gd}, \text{Tb}$ ) systems. In compositions rich with the magnetic rare earth this transition is due to the band metamagnetism (as in pure  $\text{GdMn}_2$  and  $\text{TbMn}_2$ ), whereas in yttrium-rich compositions this is a structural transition to a tetragonally distorted phase of the  $\text{YMn}_2$  type.

This conclusion can be confirmed or rejected on the basis of the results of an investigation of the behavior of the manganese subsystem in mixed intermetallics containing manganese in which there is no  $f$ - $d$  exchange interaction. With this in mind we investigated magnetic properties of mixed compounds  $\text{Y}_{1-x}\text{La}_x\text{Mn}_2$  and  $\text{Y}_{1-x}\text{Lu}_x\text{Mn}_2$  containing nonmagnetic rare earths.

#### b) Systems with nonmagnetic rare earths: $\text{Y}_{1-x}\text{Lu}_x\text{Mn}_2$ and $\text{Y}_{1-x}\text{La}_x\text{Mn}_2$

Figure 3 shows the temperature dependences of the lattice parameter of some typical compositions of the  $\text{Y}_{1-x}\text{Lu}_x\text{Mn}_2$  system. At low lutetium concentrations (up to  $x = 0.05$ ) these compounds exhibit a structural phase transition, similar to that in pure  $\text{YMn}_2$ : below 70–90 K there is an abrupt increase in the unit cell volume and tetragonal distortions appear in the crystal structure. The structural transition exhibits temperature hysteresis and at low temperatures the cubic and tetragonal phases coexist. The compositions with  $0.05 < x < 0.1$  remain cubic throughout the investigated range of temperatures and the  $a(T)$  dependence obtained for these compounds shows no anomalies.

Replacement of yttrium with lanthanum does not (in contrast to replacement with lutetium) destroy the structural phase transition and the low-temperature tetragonal

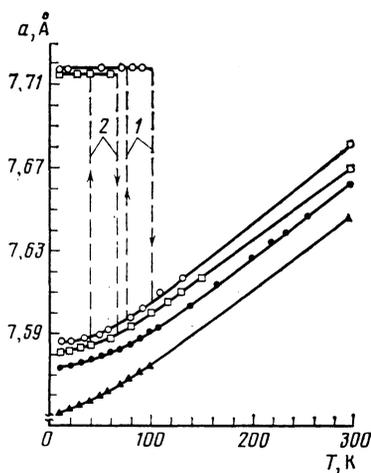


FIG. 3. Temperature dependence of the lattice parameter  $a$  of some compounds belonging to the  $\text{Y}_{1-x}\text{Lu}_x\text{Mn}_2$  system with  $x = 0$  (O), 0.03 (□), 0.05 (●), and 0.10 (Δ). The dashed lines identify the temperatures at which the low-temperature phase appears and disappears: 1)  $x = 0$ ; 2) 0.03.

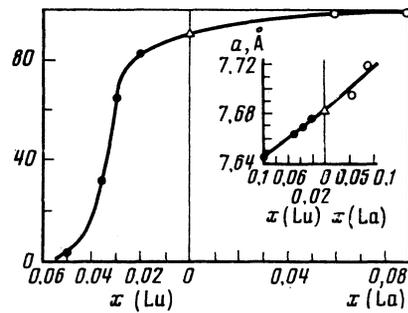


FIG. 4. Amount of the low-temperature tetragonal phase (%) at 5 K in  $\text{Y}_{1-x}\text{Lu}_x\text{Mn}_2$  (●),  $\text{Y}_{1-x}\text{La}_x\text{Mn}_2$  (○), and  $\text{YMn}_2$  (Δ) systems plotted as a function of  $x$ . The inset shows the composition dependence of the lattice parameter of these systems at 293 K.

phase exists in the  $\text{Y}_{1-x}\text{La}_x\text{Mn}_2$  system throughout the range of compositions  $0 \leq x \leq 0.09$  where we were able to prepare single-phase samples.

Figure 4 shows how the amount of the low-temperature tetragonal phase in  $\text{Y}_{1-x}\text{La}_x\text{Mn}_2$  and  $\text{Y}_{1-x}\text{Lu}_x\text{Mn}_2$  varies with  $x$  at 5 K. It is worth noting that the stability of the low-temperature phase decreases rapidly as yttrium is replaced with lutetium and in the range  $x > 0.05$  the cubic phase becomes stable at low temperatures. It should be pointed out that in the investigated range of compositions the crystal lattice parameter of the cubic phase is larger than the critical value  $a_c$  (see the inset in Fig. 4). In the system with lanthanum our experiments and those reported in Ref. 5 showed that the tetragonal phase is retained at low temperatures by all compositions characterized by  $x \leq 0.25$ .

Comparison of these results with the data for  $\text{YMn}_2$  suggests that compositions with the tetragonally distorted structure are antiferromagnetic. The magnetic state of the cubic composition was identified by neutron diffraction studies and measurements of the susceptibility of  $\text{Y}_{0.9}\text{Lu}_{0.1}\text{Mn}_2$ . The neutron diffraction data indicated that cooling of  $\text{Y}_{0.9}\text{Lu}_{0.1}\text{Mn}_2$  from 296 to 4.2 K did not alter the nature of the neutron diffractograms and there was no additional contribution to the intensity of the nuclear reflections. Figure 5 compares the neutron diffractograms of  $\text{Y}_{0.9}\text{Lu}_{0.1}\text{Mn}_2$  and antiferromagnetic  $\text{YMn}_2$  at 4.2 K. The absence of the magnetic reflections in the neutron diffracto-

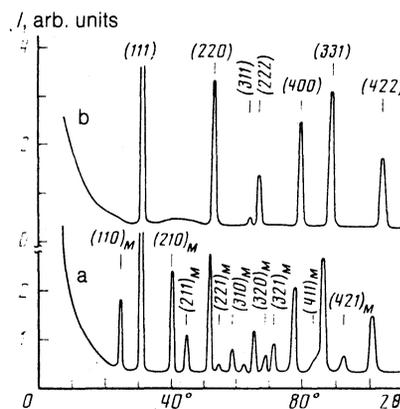


FIG. 5. Neutron diffractograms of  $\text{YMn}_2$  (a) and  $\text{Y}_{0.9}\text{Lu}_{0.1}\text{Mn}_2$  (b) at 4.2 K. The magnetic reflections are identified by the subscript  $M$ .

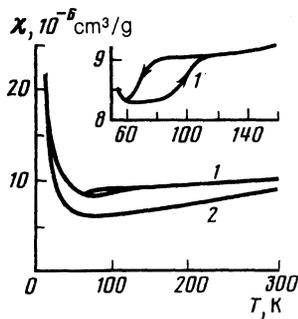


FIG. 6. Temperature dependences of the magnetic susceptibility of  $Y_{1-x}Lu_xMn_2$  compounds with  $x = 0$  (1) and  $x = 0.1$  (2). The inset shows the dependence  $\chi(T)$  for  $YMn_2$  in the phase transition region.

gram of  $Y_{0.9}Lu_{0.1}Mn_2$  indicates that this compound is paramagnetic in the investigated range of temperatures.

These neutron diffraction results are supported by measurements of the paramagnetic susceptibility of  $Y_{0.9}Lu_{0.1}Mn_2$ . Figure 6 shows the temperature dependence  $\chi(T)$  obtained for this compound and for  $YMn_2$ . Clearly, the susceptibilities of both intermetallics are similar. It follows that the density of states at the Fermi level changes only slightly as a result of partial replacement of yttrium with lutetium. Moreover, the temperature dependence  $\chi(T)$  is qualitatively similar with the exception that  $Y_{0.9}Lu_{0.1}Mn_2$ , in contrast to  $YMn_2$ , exhibits no susceptibility anomalies near 90 K (such an anomaly is known to be due to the antiferromagnetic ordering).<sup>8</sup>

The coexistence of the low- and high-temperature phases (Fig. 3) and gradual variation of the relative amounts of these phases with temperature (i.e., the heterophase nature of a sample) broadens the susceptibility anomaly of  $YMn_2$  to a wide temperature interval (inset in Fig. 6). This heterophase nature of the sample is responsible for the gradual change in any macroscopic properties of  $YMn_2$ , such as its thermal expansion,<sup>14</sup> specific heat,<sup>15</sup> etc. in the vicinity of the structural transition.

## DISCUSSION OF RESULTS

Our experiments demonstrate that the low-temperature tetragonally distorted phase of  $YMn_2$  becomes destabilized in most cases at a relatively low concentration of the rare earth replacing yttrium; this happens when the lattice parameter is still considerably greater than the critical value  $a_c$ . This is demonstrated clearly in Fig. 7 which shows the composition dependences of the volume anomaly, which appears as a result of the transition from the cubic to the tetragonal phase at 5 K in the  $Tb_{1-x}Y_xMn_2$  (Ref. 6),  $Gd_{1-x}Y_xMn_2$ ,  $Y_{1-x}Lu_xMn_2$ , and  $La_{1-x}Y_xMn_2$  systems investigated. This figure includes also the data<sup>16</sup> for the  $Y_{1-x}Sc_xMn_2$  system where replacement of yttrium with the 3d metal scandium also destabilizes the tetragonally distorted phase. In mixed compositions this phase disappears when the lattice parameter is both reduced (due to replacement with Tb, Lu, or Sc) and increased (replacement with Gd).

All the points mentioned above demonstrate that the transition to the tetragonally distorted phase with an antiferromagnetic manganese subsystem is of special nature and it occurs not in all  $RMn_2$  compounds, but only in  $YMn_2$  and in yttrium-rich mixed  $(Y,R)Mn_2$  compounds. This transition is structural and in many respects similar to the transition

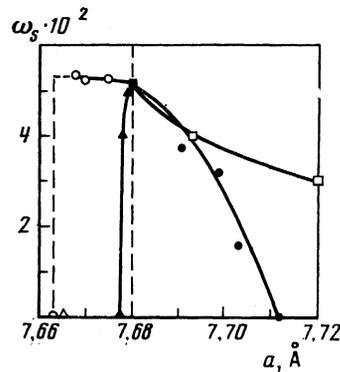


FIG. 7. Dependence of the volume anomaly  $\omega_s$  at 5 K on the lattice parameter of  $YMn_2$  (■) and  $(Y,R)Mn_2$  with  $R = La$  (□),  $Lu$  (○),  $Tb$  (△),  $Gd$  (●), and  $Sc$  (△). The critical concentrations  $x_c$  responsible for stabilization of the cubic phase in  $(Y,R)Mn_2$  systems are as follows: 0.05 for  $R = Lu$ ; 0.1 for  $Tb$  (Ref. 6); 0.4 for  $Gd$ ; 0.02 for  $Sc$  (Ref. 16).

observed in  $UMn_2$  at a temperature below  $\approx 220$  K (Ref. 17).

The nature of the structural instability of the cubic  $YMn_2$  phase at low temperatures is not yet clear. It is obviously due to fine details of the energy spectrum of the  $d$  electrons. Stabilization of the cubic phase by partial replacement of yttrium with rare earths (or scandium) demonstrates differences between the band structure of the  $4d$  electrons of yttrium in  $YMn_2$  and the  $5d$  electrons of rare earths in  $RMn_2$  (and the  $3d$  electrons of scandium in  $ScMn_2$ ). In particular, the high stability of the tetragonal phase in the  $(Y,La)Mn_2$  system demonstrates that the nature of changes in the energy state of the  $d$  electrons due to replacement of yttrium with lanthanum is different from changes in other mixed systems based on  $YMn_2$  (the difference between the  $d$  band of lanthanum and the  $d$  band of other rare earths is demonstrated in many ways and in particular by the fact that intermetallic compounds do not form<sup>18</sup> in the  $Mn-La$  double system).

It therefore follows that the rigid  $d$  band model is inappropriate for a combined discussion of the magnetic properties of the manganese subsystem in the low-temperature phase of  $YMn_2$  and in cubic  $RMn_2$  compounds containing magnetic rare earths. However, in the first approximation, we can assume that the structures of the  $d$  bands are the same for pure  $RMn_2$  compounds and for  $(R,Y)Mn_2$  systems rich in a rare earth, which exhibit no low-temperature structural transition.

We shall now use the band metamagnetism model to consider the magnetic behavior of the manganese subsystem in pure  $RMn_2$  and in mixed  $(R,Y)Mn_2$  and  $(R,Lu)Mn_2$  compounds as a function of the atomic number of the rare earth. The  $H_{R-Mn}$  exchange field due to the rare-earth subsystem acting on the manganese subsystem can be represented as follows for mixed systems:

$$H_{R-Mn} = (1-x)\lambda_{R-Mn}M_R = (1-x)I_{R-Mn}(g_R-1) \cdot J_R \mu_B, \quad (3)$$

where  $\lambda_{R-Mn}$  is the molecular field coefficient;  $M_R$  and  $1-x$  are the magnetic moment and the concentration of the magnetic rare earth, respectively. It is assumed in Eq. (3) that the spin exchange parameter  $I_{R-Mn}$  is independent of the atomic moment of the rare earth (as is true also of  $RCO_2$  compounds<sup>19</sup>).

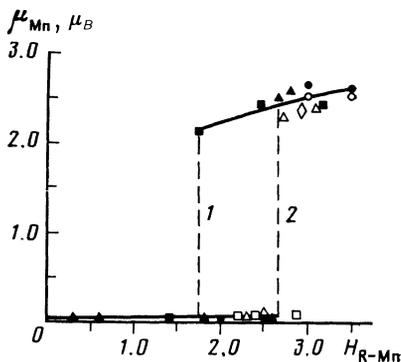


FIG. 8. Dependence of the magnetic moment of manganese on the field  $H_{R-Mn}$  (arb. units) in  $RMn_2$  compounds with  $R = Dd, Tb, Dy, Ho$  (●, Ref. 1; ○, Ref. 2), and in mixed systems  $(Tb, Y)Mn_2$  (△, Ref. 6; ◇, Ref. 12),  $(Tb, Lu)Mn_2$  (□, Ref. 12),  $(Gd, Er)Mn_2$  (△, Ref. 12),  $(Gd, Y)Mn_2$  (■). 1)  $H_c$  for the  $Gd_{1-x}Y_xMn_2$  system; 2)  $H_c$  for the other systems.

Figure 8 shows the dependence of the magnetic moment of manganese on the molecular field  $H_{R-Mn}$  in pure  $RMn_2$  compounds ( $R = Gd, Tb, Dy, Ho$ ) and in mixed  $(Tb, Y)Mn_2$ ,  $(Gd, Y)Mn_2$ ,  $(Tb, Lu)Mn_2$ , and  $(Gd, Er)Mn_2$  systems, deduced from the NMR spectra<sup>12</sup> and from our own x-ray diffraction data applying Eq. (1) (where the coefficient  $k$  is found from the values of  $\omega_s$  and  $\mu_{Mn}$  for  $GdMn_2$ ). In this figure we give the results solely for the cubic compositions with the  $MgCu_2$ -type structure, for which the band structure of the  $d$  subsystem can be regarded as almost the same. We can see that the manganese subsystem is magnetically ordered in a high exchange field  $H_{R-Mn}$  and becomes disordered abruptly when this field is reduced. The dependence  $\mu_{Mn}(H_{R-Mn})$  demonstrates the metamagnetic nature of this curve and the critical field of the metamagnetic transition lies between  $H_{Tb-Mn}$  and  $H_{Dy-Mn}$  (with the exception of the  $Gd_{1-x}Y_xMn_2$  system for which it is somewhat less probably because  $I_{R-Mn}$  for this system differs from the corresponding parameter of the other  $RMn_2$  compounds).

It therefore follows that the band metamagnetism model is capable (in contrast to the model of the critical Mn-Mn distance) of explaining the magnetic properties of the manganese subsystem in pure and mixed  $RMn_2$  compounds with the cubic crystal lattice of the  $MgCu_2$  type. It should be pointed out that band metamagnetism in  $RMn_2$  intermetallics can appear, consistent with the calculated energy band structure of cubic  $YMn_2$  in the paramagnetic state. According to Ref. 20, the Fermi level of this compound lies near a minimum of the density of states  $N(\epsilon)$ . This ensures an increase in  $N(\epsilon_F)$  in an external magnetic field and the appearance of the magnetic order in the  $d$ -electron system when a certain critical value of the field  $H_c$  is reached.

## CONCLUSIONS

The above analysis shows that the nature of the band subsystem magnetism in  $RMn_2$  and  $RCO_2$  Laves phases is similar: it is due to a metamagnetic transition in the  $f$ - $d$  exchange field. However, the external magnetic properties of these two classes of compounds are very different. In our opinion, this is due to the following factors. Firstly, in the case of  $YMn_2$  the low-temperature structural instability gives rise to the antiferromagnetic ordering, whereas  $YCO_2$  is a band paramagnet up to 4.2 K. Secondly, the intrasublattice

$f$ - $f$  exchange interaction in  $RCO_2$  is positive, whereas the intersublattice  $f$ - $d$  interaction is negative.<sup>19</sup> In  $TbMn_2$  both interactions are negative.<sup>21</sup> Thirdly, the parameter  $I_{R-Co}$  of  $RCO_2$  is greater than  $I_{R-R}$ , whereas in the case of  $RMn_2$  the parameter  $I_{R-Mn}$  clearly does not exceed  $I_{R-R}$ . The competition between two comparable negative interactions is possibly the reason for the complex nonlinear magnetic structures of intermetallics  $GdMn_2$  and  $TbMn_2$  containing a magnetically ordered manganese subsystem.

Summarizing the results we can say that in discussing the magnetic properties of  $RMn_2$  intermetallics it is desirable to divide these compounds (because of the differences between the structure of the  $d$  band) into three groups: 1) cubic  $RMn_2$  with the  $MgCu_2$ -type structure, in which the manganese subsystem has the band metamagnetic properties; 2) hexagonal  $RMn_2$  with the  $MgZn_2$  structure, the magnetism of which is not yet clear; 3)  $YMn_2$  which undergoes a low-temperature structural transition to a tetragonally distorted phase.

<sup>1</sup>Note that for  $H_{R-Mn} < H_c$ , the paramagnetic  $d$ -electron system has a definite paramagnetic moment induced by the  $f$ - $d$  exchange fields, whereas for  $H_{R-Mn} > H_c$  the magnetic moment of the  $d$  electrons is due to the combined effects of the  $d$ - $d$  and  $f$ - $d$  exchange fields.<sup>4</sup>

<sup>2</sup>Note that this result is in conflict with the NMR data given in Ref. 12, according to which at 4.2 K the moment of manganese differs from zero in  $Gd_{0.4}Y_{0.6}Mn_2$ . This conflict may be due to the fact that the range of compositions where the manganese subsystem is paramagnetic is very narrow (Fig. 2) and the samples prepared by various methods may have very different magnetic properties near the boundaries of this region. This question requires further experimental study.

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