Pressure-induced magnetic transitions in the frustrated Laves compound $GdMn_2$

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We studied the magnetic and crystal structure of the magnetically unstable compound $GdMn_2$ using neutron diffraction and x-ray diffraction at ambient pressure and applied pressure up to 4.4 GPa (neutron) and 16 GPa (x ray). At ambient pressure, $GdMn_2$ shows a short-range antiferromagnetic order with a ferromagnetic component appearing at low temperature. As pressure increases, the Mn moments transform from an intrinsic to an induced state around 1.5 GPa and then vanish completely. This transformation yields a suppression of the short-range antiferromagnetic correlations imposed by the topologically frustrated Mn sublattice. We show that in the pressure range $2<P<4$ GPa spin fluctuations on the Mn sites prevent a complete magnetic order in the Gd sublattice. Finally, above 4 GPa, the system recovers a purely ferromagnetic disorder-free magnetic state. We show that $GdMn_2$ exhibit a very intriguing and, in some sense, unique interplay between the *f* and *d* magnetisms.

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I. INTRODUCTION

The magnetic Laves phases RMn_2 (R=rare earth) are very intriguing systems. In these compounds, the Mn ions are close to the instability limit between localized and itinerant magnetic states.[1] The magnetic state is controlled by the value of the first-neighbor Mn-Mn distance ($d_{Mn\text{-}Mn}$). In the compounds with lighter rare earth having large ionic radii (R=Pr,Nd,Tb,Gd,Y), the Mn ions carry intrinsic magnetic moments; whereas in the compounds with heavier rare earth having smaller ionic radii (R=Er,Tm,Lu), the Mn sublattice is nonmagnetic. On the other hand, the Mn sublattice of corner-sharing tetrahedra is topologically frustrated, i.e., no spin arrangement can satisfy all first-neighbor antiferromagnetic interactions and the magnetic ground state is degenerated.[2–4] The interplay between the topological frustration and the magnetic instability results in very unusual physical phenomena. The system might release the frustration through a structural distortion (YMn_2), or by canceling magnetic moments at some selected Mn sites ($TbMn_2$, $HoMn_2$), or it could remain in a spin-liquid state down to T=0 K [$Y(Mn,Sc)_2$]. If R is magnetic, the interaction between the R and Mn sublattices yields other interesting phenomena. While *d-d* and *f-d* magnetic interactions are expected to be antiferromagnetic (i.e., negative), the *f-f* exchange is expected to be ferromagnetic (i.e., positive), so the magnetic ground state is determined by a delicate balance of magnetic interactions having different signs. In some cases, the magnetic rare earth can stabilize magnetism in the Mn sublattice. It is believed that in $HoMn_2$ and $DyMn_2$, the local exchange fields from the rare-earth sublattice induce magnetic moments on the Mn sites.[5,6] Contrarily, in $TbMn_2$ the intrinsic Mn magnetic moments impose magnetic order in the Tb sublattice.[1]

Among all RMn_2 compounds, $GdMn_2$ shows the most intriguing interplay of the *f* and *d* magnetisms. In $GdMn_2$, the $d_{Mn\text{-}Mn}$ distance is slightly above the critical value so the Mn ions are expected to carry localized moments, although being very close to the magnetic instability. Among the 4*f* ions, Gd^{3+} carries the largest spin (S=7/2). Therefore, in $GdMn_2$, the *f-f* and *f-d* contributions to the magnetic free energy can be comparable to the *d-d* contribution and can influence the ground state in both *d* and *f* sublattices. Despite the obvious interest in the magnetic properties of $GdMn_2$, information on magnetic ordering in $GdMn_2$ is limited. Magnetization and thermal expansion measurements show two anomalies at ~110 K and ~40 K.[1,7,8] While the transition at 110 K was identified as a Néel transition to an antiferromagnetically ordered structure, a nonzero magnetization below 40 K suggests a ferromagnetic or ferrimagnetic structure at a low temperature. The anomaly in the magnetic properties at 110 K is accompanied by a considerable volume expansion, which is a characteristic feature for ordering of intrinsic *d* moments. It was suggested that the two temperatures (denoted as T_N and T_C) correspond to the magnetic orderings in the Mn and Gd sublattices, respectively, which occur independently.[7] Nevertheless, specific heat, μSR, and Mössbauer measurements suggest that both sublattices order simultaneously at ~110 K and the transition at 40 K is more likely related with a reorientation of magnetic moments.[9–12] Unfortunately, neutron diffraction, the only direct method to study microscopic spin arrangement, is difficult in the case of $GdMn_2$ because of the huge neutron absorption in Gd. Available neutron diffraction data, obtained on a highly absorbing sample, provide limited information on magnetic structure.[13] They confirm the simultaneous ordering of the Mn and Gd magnetic moments and suggest a noncollinear antiferromagnetic structure, similar to that found in $TbMn_2$. In contradiction with other probes, no anomaly was found at T=40 K and no ferromagnetic component was observed at the lowest temperature. So the "40 K anomaly" remained a mystery.

The huge sensitivity of the Mn magnetism to interatomic distances provides an opportunity to study the respective

roles of the f and d magnetisms by changing the relative strength of the two sublattices in experiments under applied pressure. In $GdMn_2$, pressure is expected to destroy intrinsic Mn moments, as was found in YMn_2,[14] whereas the f ions carrying well-localized moments are expected to be relatively insensitive to pressure. Resistivity,[15,16] magnetization,[7–20] and thermal expansion measurements[21,22] allowed us to trace the pressure behavior of T_N and T_C. Whereas T_C increases with pressure, T_N decreases rapidly and disappears completely at the critical pressure $P_c \sim 1.5$ GPa. Therefore, it was argued that pressure stabilizes ferromagnetic order in $GdMn_2$. Although there was no direct proof, this conclusion was supported by Mössbauer measurements.[23] The negative volume anomaly at T_N persists under pressure, suggesting that T_N is associated with the ordering of intrinsic Mn moments in the whole pressure range below P_c. Assuming this scenario, in the pressure range $0.9 < P < 1.5$ GPa the intrinsic d moments order at *lower* temperatures than the f moments, which is a unique case in all RMn_2 compounds. Another intriguing feature is the huge increase of T_C under pressure. At $P=3$ GPa T_C reaches ~ 155 K and becomes considerably higher than the temperature of magnetic ordering at ambient pressure (110 K), therefore one comes to the paradoxical conclusion that the *suppression* of magnetic moments on some sites results in an *increase* in the temperature of magnetic ordering.

Despite numerous high-pressure studies, there was no direct information on the magnetic structure in $GdMn_2$ under pressure. Nothing was known about the pressure dependence of microscopic magnetic moments on the Mn and Gd sites, and the exact spin arrangement in the pressure-induced magnetic phase at $P > P_c$ was not known. Only neutron diffraction can answer the above questions. Below, we report a detailed neutron diffraction study of $GdMn_2$ at ambient and high pressures. The neutron experiments were carried out under pressures up to 4.4 GPa, far beyond the pressure range where the magnetic transformations are expected to take place. Additional information on the crystal structure under pressure was obtained in x-ray experiments.

II. EXPERIMENTAL DETAILS

Neutron diffraction measurements were carried out on the powder neutron diffractometer G6.1 "MICRO" installed at the ORPHEE reactor of the Laboratoire Léon Brillouin. We used nonabsorbing isotopically enriched $^{160}GdMn_2$. A sapphire anvil cell was used to generate pressure.[24] Neutrons were focused to the sample by a special focusing system. Temperature was varied between 1.5 and 300 K. In order to characterize the crystal structure under pressure, x-ray diffraction experiments were performed at the ID30 beamline at the European Synchrotron Radiation Facility. In this case, we used a diamond-anvil cell and experiments were made at room temperature. In both cases (neutron and x ray), the ruby fluorescence was used to measure pressure with a typical error of ±0.1 GPa. Information on the magnetic structure and values of ordered magnetic moments were derived from the positions of magnetic peaks and their intensities using the FULLPROF program.[25]

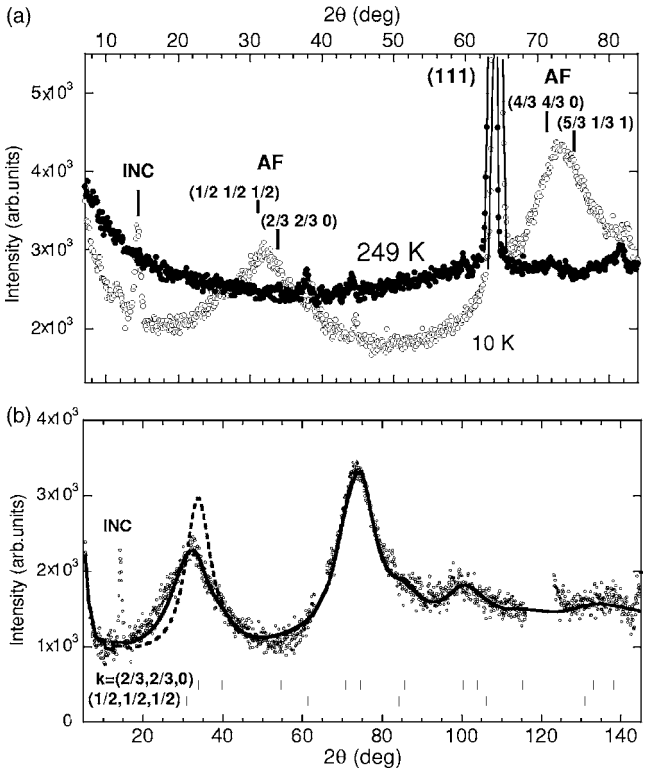

FIG. 1. (a) Neutron diffraction patterns of $GdMn_2$ measured at temperatures above (249 K, filled circles) and below (10 K, open circles) the temperatures of magnetic transitions at ambient pressure. Neutron wavelength is 4.74 Å. INC and AF denote incommensurate and commensurate antiferromagnetic peaks, respectively. (b) The difference between the low- and high-temperature spectra. The dashed and solid lines correspond to fits of the diffraction data using one propagation vector ($\mathbf{k}=2/3,2/3,0$) or two propagations vectors ($\mathbf{k}=2/3,2/3,0$ and $1/2,1/2,1/2$), respectively. Only the positions and widths of the antiferromagnetic peaks were fitted, whereas the intensities were free parameters. Tics at the bottom of the figure show calculated positions of the magnetic peaks.

III. EXPERIMENTAL RESULTS

In Fig. 1 we show the neutron diffraction patterns of $GdMn_2$ measured at ambient pressure. Below 110 K we observed additional superstructural peaks, which are attributed to the formation of an antiferromagnetic order in the Mn and Gd sublattices. The magnetic peaks are much broader than the structural ones, which indicates a short-range magnetic order with correlation length of 30–50 Å. In agreement with Ref. 13, the largest fraction of the magnetic scattering is attributed to the propagation vector $(2/3,2/3,0)$. Nevertheless, we could not describe *all* magnetic scattering with only one propagation vector. An additional antiferromagnetic modulation with the propagation vector $(1/2,1/2,1/2)$ is required to describe the magnetic peaks [Fig. 1(b)]. In addition to the commensurate magnetic peaks, we observed a small narrow incommensurate peak at low angles ($2\theta \sim 15°$). Both commensurate and incommensurate components appear at the same temperature 110 K [Fig. 2(a)]. At $T=40$ K, we observed another anomaly in the neutron diffraction patterns. While the antiferromagnetic peaks remain

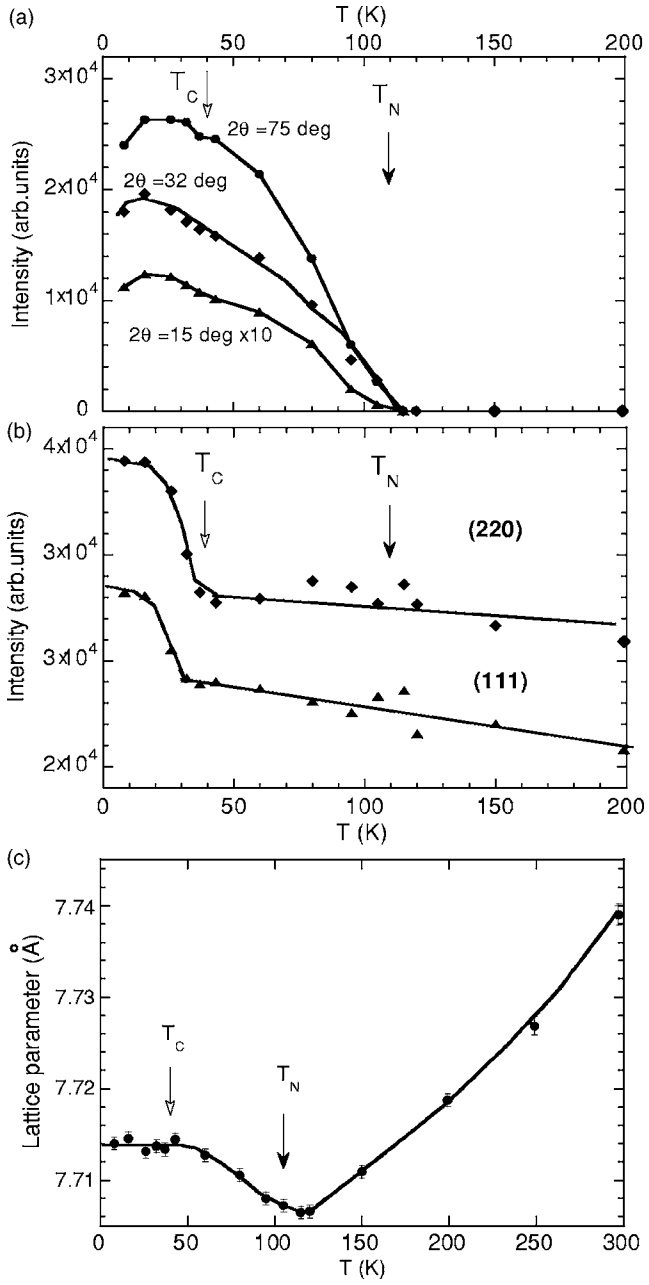

FIG. 2. (a) Temperature dependencies of the antiferromagnetic (circles and crosses) and the incommensurate modulation (triangles) peaks of $GdMn_2$ at ambient pressure. (b) Temperature dependencies of the structural peaks (111) and (220), which show a ferrimagnetic component below 40 K. (c) Lattice parameter versus temperature at ambient pressure.

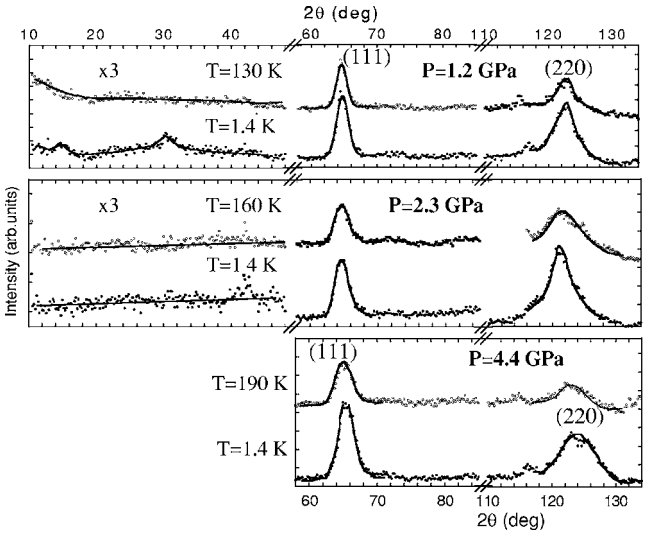

FIG. 3. Neutron scattering in $GdMn_2$ measured at different pressures and temperatures. For convenience, the spectra measured at the high and low temperatures are shifted along the y axis. As pressure increases, short-range antiferromagnetic correlations at small scattering angles disappear, whereas low temperature contributions to the structural peaks, located at large scattering angles, increase.

almost unchanged except a tiny decrease at $T=10$ K [Fig. 2(a)], additional scattering appears at the position of structural peaks (111) and (220) [Fig. 2(b)], indicating a presence of a ferromagnetic or ferrimagnetic component in the magnetic structure. Contrarily to the antiferromagnetic peaks, these contributions are relatively narrow, although slightly broader than the structural contributions. Their widths correspond to a correlation length of 300–500 Å. The finite correlation length in the ferro(ferri)magnetic component of the magnetic structure is also evidenced from the appearance of a small-angle scattering ($2\theta < 10°$) at low temperature (Fig. 1). Refinement of the magnetic intensities yields ferromagnetic components in both Gd and Mn sublattices. The ferromagnetic component on the Gd sites is estimated to 2.5 μ_B at $T=10$ K. The ferromagnetic component on the Mn sites is antiparallel to the ferromagnetic component on the Gd sites and estimated to 0.5 μ_B at 10 K. These values are considerably lower than the expected full Gd and Mn moments (7 and 2.5 μ_B, respectively). We conclude that the antiferromagnetic component dominates in the magnetic structure even at the lowest temperature and the transition at 40 K should be attributed to a partial reorientation of the magnetic moments. In agreement with the previous studies, the magnetic ordering at 110 K is accompanied by a considerable volume expansion (0.6%), whereas the reorientation at $T=40$ K has no obvious effect on lattice parameter [Fig. 2(c)].

Under pressure, we observed a drastic change in the magnetic scattering (Figs. 3–5). At $P=1.2$ GPa, as temperature decreases, a collinear ferrimagnetic order appears first at 110 K, whereas the antiferromagnetic correlations develop at lower temperatures $T<80$ K [Fig. 4(a)]. This agrees with the information obtained by indirect probes. Nevertheless, while Ref. 22 suggests that in the temperature range $T_N<T<T_C$ the Mn sublattice is nonmagnetic, we detect a nonzero magnetization in the Mn sublattice in this temperature range [Fig. 4(b)]. In agreement with indirect probes, T_C increases and reaches 138 and 155 K at $P=2.3$ and 4.4 GPa respectively [Fig. 5(a)]. At $P=2.3$ GPa the antiferromagnetic modulation disappears completely together with the incommensurate magnetic peak, and the magnetic structure can be described as a collinear ferrimagnet with magnetic moments of 4.6 μ_B and 0.35 μ_B on the Gd and Mn sites, respectively. As at $P=0$, the Gd and Mn moments are antiparallel. From our neutron data, we cannot choose between a small moment of

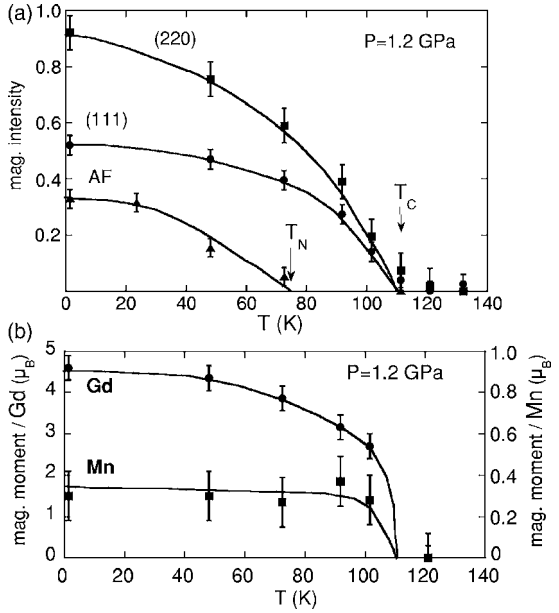

FIG. 4. (a) Integrated magnetic intensities of the ferrimagnetic peaks (111), (220), and the AF peak versus temperature at $P=1.2$ GPa. (b) The ferrimagnetic component of the magnetic moments on the Gd and Mn sites at $P=1.2$ GPa versus temperature.

0.35 μ_B uniformly distributed among all Mn sites, or a larger moment of 1.4 μ_B concentrated on 1/4 of the Mn sites, as was found in $DyMn_2$ and $HoMn_2$ at ambient pressure.[5,6] Finally, at 4.4 GPa we did not observe any magnetic moments on the Mn sites, and the ordered Gd moment is estimated to be $6.6(4)\mu_B$, which is very close to the free-ion value [Fig. 5(b)]. We conclude that at this pressure, $GdMn_2$ becomes a

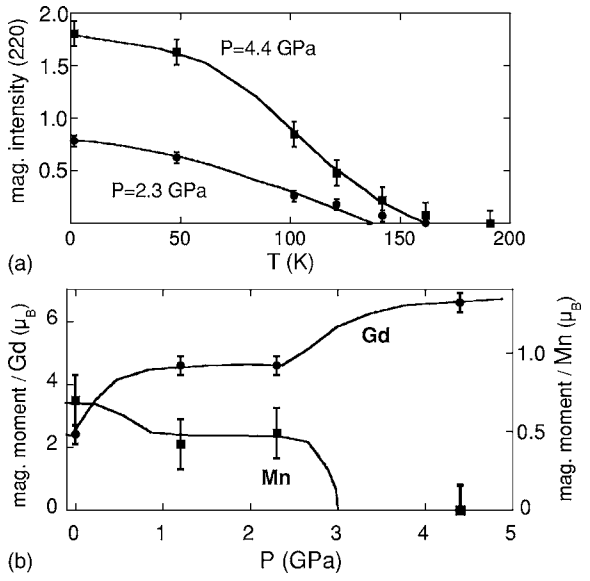

FIG. 5. (a) Integrated magnetic intensity of (220) peak at $P=2.3$ and 4.4 GPa versus temperature. The magnetic intensity was normalized to the intensity of the structural peak (220) at $T>T_C$. (b) Ferrimagnetic components of the magnetic moments on the Gd and Mn sites at different pressures and the lowest temperatures (10 K at ambient pressure and 1.5 K at high pressures).

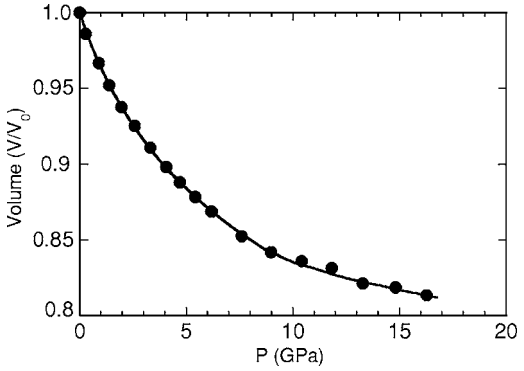

FIG. 6. X-ray data on the relative unit cell volume in $GdMn_2$ at $T=300$ K and high pressures. The solid line is fit by the Murnaghan equation.

simple collinear ferromagnet with a nonmagnetic Mn sublattice.

The transition from the intrinsic to an induced or a nonmagnetic state could be accompanied by a significant decrease in the unit cell volume. In RMn_2, such anomaly can reach up to −5.5% (YMn_2).[26] In Fig. 6, we show x-ray data on the unit cell volume in $GdMn_2$ under pressures up to 16 GPa, measured at room temperature. We did not observe any sharp anomaly on the $V(P)$ curve, in agreement with recently published data.[27] No evidence was found for a structural transition (C15→C14) as suggested in Ref. 22. We note the very high compressibility in the pressure range $P<10$ GPa. The bulk modulus B_0 is three to four times lower than the value expected in the intermetallic f-d compounds ($B_0=22.9$ GPa, compared to typical values of 50–100 GPa) and its pressure derivative is abnormally high ($B'=8.5$, compared to the typical values of 3–5). This abnormal pressure behavior could be naturally explained by the influence of spin fluctuations in the Mn sublattice. As it was shown in YMn_2,[1] the spin fluctuations enlarge the unit cell volume. Under pressure, as Mn moments vanish and the system moves out of the range of the magnetic instability, spin fluctuations gradually decrease, which results in an additional decrease of the unit cell volume and an abnormally high compressibility. Abnormal values of B_0 and B_1 (19 GPa and 15.2, respectively) were also found in the other magnetically unstable Laves compound YMn_2.[28]

IV. DISCUSSION

Now we can plot a general picture of magnetic phenomena in $GdMn_2$ at different pressures. In Fig. 7 we show the P-T magnetic phase diagram of $GdMn_2$. At low pressures (<1 GPa), the d-d interactions dominate and impose the antiferromagnetic order in both Gd and Mn sublattices. At the lowest temperatures, the f-f magnetic exchange becomes sufficiently strong to tilt the magnetic moments and induce a ferromagnetic component. The reorientation occurs in both Gd and Mn sublattices, which are coupled through the f-d magnetic exchange. Under pressure, the intrinsic Mn moments decrease and the relative contribution of the d-d exchange to the magnetic free energy decreases. At P

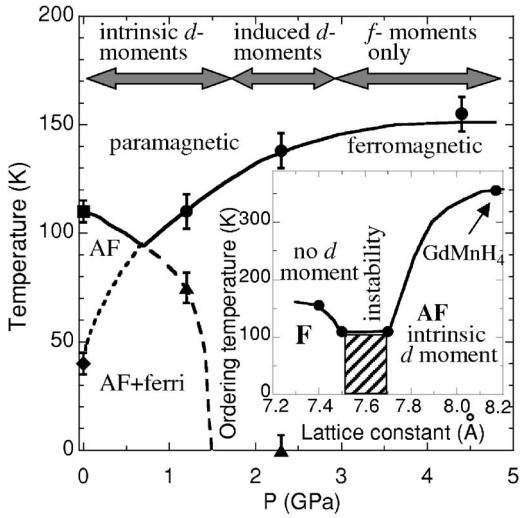

FIG. 7. Magnetic P-T phase diagram derived from neutron data. Square: antiferromagnetic ordering occurring simultaneously in the Mn and Gd sublattices; triangles: AF ordering occurring independently in the Mn sublattice; circles: ferro(ferri)magnetic ordering; rhomb: reorientation of magnetic moments yielding a ferromagnetic component in the Mn and Gd sublattices. Solid and dashed lines show the transition to the paramagnetic state and the low-temperature magnetic transitions, respectively. In the inset: the temperature of transition to the paramagnetic state in $GdMn_2$ (ambient and high pressures, neutron data) and in $GdMn_2H_{4.4}$ (ambient pressure, data from Ref. 31) versus lattice constant.

$=1.2$ GPa, the situation is about opposite to that at ambient pressure. The f-f interactions determine the magnetic order at high-temperature $T_N<T<T_C$, whereas the d-d interactions induce an antiferromagnetic component at the lowest temperature $T<T_N$. Bringing together the neutron data and the data on thermal expansion,[22] one can conclude that the intrinsic magnetic ordering in the Mn sublattice develops in the temperature range $T<T_N$. Nevertheless, even at higher temperatures $T_N<T<T_C$ the f-d exchange is sufficiently strong to induce a small ferrimagnetic component on the Mn sites. As pressure increases further, the Mn intrinsic moments vanish completely and $GdMn_2$ becomes a collinear ferrimagnet in the whole temperature range below the magnetic ordering temperature. $P=2.3$ GPa is an upper limit for the pressure P_c of suppression of intrinsic Mn moments. At 2.3 GPa we still detect a small magnetic moment on the Mn sites, induced by the exchange field of the Gd sublattice. At $P>4$ GPa the magnetic evolution is completed and the system becomes a casual ferromagnet with no moments on the Mn sites.

The high-pressure data provide crucial information on the microscopic origin of the short-range and incommensurate magnetic orderings in the RMn_2 compounds. Besides $GdMn_2$, short-range antiferromagnetic ordering was observed in the doped compound[29] $Ho(Mn_{0.9}Al_{0.1})_2$ and the Laves hydride $Tb(Mn_{0.88}Al_{0.12})_2H_{1.04}$.[30] It was shown that the short-range ordered state can be easily suppressed by pressure. One can argue that the chemical doping could stabilize a short-range magnetic order. We observed a short-range antiferromagnetic structure in a nondoped Laves phase. Our data provide a direct proof that the short-range order originates from the specific topological and magnetic properties of the Mn sublattice. Under pressure, as the intrinsic Mn moments disappear, the system recovers long-range commensurate magnetic order.

Other interesting features of $GdMn_2$ are the pressure dependences of the ordered magnetic moments and the magnetic ordering temperature in the Gd sublattice. At low pressures, the Gd sublattice is governed by the frustrated short-range ordered Mn sublattice. Therefore, one can naturally expect that some component of the magnetic moments on the Gd sites would be disordered or short-range ordered down to the lowest temperature, and long-range ordered moments in the Gd sublattice would be lower than the free-ion value. Nevertheless, while at $P=2.3$ GPa, the localized Mn moments vanish completely and we do not observe any sign of magnetic disorder, the value of the ordered moment on the Gd sites is still much lower than the free-ion value (4.6 compared to $7\mu_B$). The rapid increase in the values of the ordered Gd moments accompanied by an increase in T_C takes place in the pressure range $2<P<4$ GPa, where no essential changes in the magnetic structure were found. What could be the microscopic origin for the partial suppression of the ordered moments on the Gd sites in the pressure range $2<P<4$ GPa? We suggest that spin fluctuations on the Mn sites, which persist above 2.3 GPa, influence the Gd moments through f-d magnetic exchange and prevent a complete magnetic ordering in the Gd sublattice. This idea is supported by the x-ray data, which show an abnormal compressibility in the whole pressure range <5 GPa.

$GdMn_2$ is a very interesting case where a magnetically unstable sublattice could influence magnetic order in such a way that the partial suppression of magnetism results not in a *decrease*, but in an *increase* of the net magnetization and the ordering temperature. When plotting the magnetic ordering temperature versus lattice parameter in $GdMn_2$ and $GdMn_2H_{4.4}$ (in the hydride, the lattice parameter is about 7% larger than in $GdMn_2$)[31] (Fig. 5), one can see that the ordering temperature passes through a minimum. This minimum corresponds to the magnetic instability in the Mn sublattice, whereas the left and the right parts of the graph correspond to the stable antiferromagnetic "d driven" and ferromagnetic "f driven" magnetic states, respectively.

In conclusion, we report a consistent picture of a very intriguing interplay between the f- and d-magnetic sublattices in the frustrated Laves compound $GdMn_2$.

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