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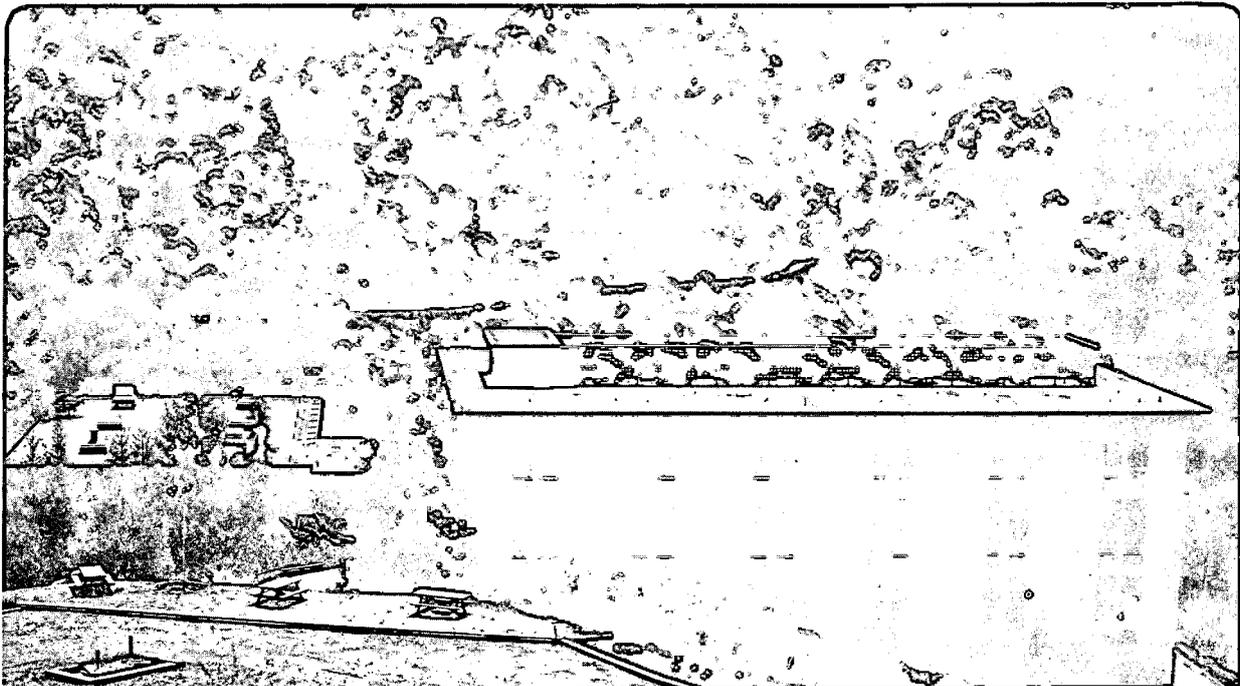
Materials Sciences Division

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Low-Temperature Specific Heat of YMn_2 in the Paramagnetic and Antiferromagnetic Phases

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The low-temperature specific heat of YMn_2 has been measured at applied pressures of 0 to 7.7 kbar. A paramagnetic state is stabilized for moderate values of the applied pressure (of the order of 1.6 kbar). A large linear term in the specific heat, which decreases regularly with increasing pressure, is observed in this phase. It is ascribed to giant spin fluctuations associated with a magnetic-non magnetic instability and a strong geometrical spin frustration.

1. Introduction

Interesting magnetic properties occur in the RMn_2 intermetallic compounds ($R = Y, Sc, Th$ or a Lanthanide) associated with a combined effect of a magnetic-non magnetic (M-NM) instability and a strong magnetic frustration of the Mn itinerant-electron antiferromagnetism. It has been suggested that a critical spacing exists between the Mn ions below which Mn is not magnetic and above which a large Mn moment (about $2.6 \mu_B$) is stabilized¹. As a result of the M-NM instability, there is a large thermal expansion (which reflects the presence of large-amplitude low-energy spin fluctuations) in the paramagnetic phase, and a large magnetovolume effect (which reflects a substantial increase in the Mn magnetic moment) at the ordering temperature. The RMn_2 compounds crystallize in either the C14 hexagonal or the C15 cubic Laves phases. In these structures, the Mn ions are located at the vertices of regular tetrahedra which are connected by alternating shared vertices and shared bases in the hexagonal phase, and by shared vertices in the cubic phase. In both cases, a strong geometrical spin frustration would result for antiferromagnetism. In particular, with first neighbor exchange interactions only, no magnetic order could set up as a result of infinite spin configurational degeneracy.

Within the RMn_2 series, YMn_2 , which crystallizes in the cubic C15 phase and in which Y is not magnetic, shows the largest magnetovolume effect. It orders in an antiferromagnetic structure of the first kind for a fcc cubic lattice with a long wavelength distorted helical component². The transition is first order, characterized by a large thermal hysteresis depending on the sample and is accompanied by a 5% increase of the volume¹. A large thermal expansion coefficient is measured in the paramagnetic phase ($\beta = (1/V)(\partial V/\partial T)_{PH} \approx 150 \cdot 10^{-6} K^{-1}$). A high sensitivity of this magnetic state, with respect to an applied hydrostatic pressure or to a chemical pressure, is observed. With hydrostatic pressure, the Néel temperature decreases³ at a rate as large as $\partial T_N/\partial p \approx -35 K/kbar$. A small substitution of Fe, Co or Al for Mn or of Sc for Y, destabilizes the magnetic order⁴. Striking features characterize the magnetic behavior of the $Y(Sc)Mn_2$ substituted compounds. It has been shown that the linear contribution, γT , to the specific heat of the compound with 3% of Sc is enormously large⁵ for a 3d metal : γ reaches a value of about $140 mJ.K^{-2}mol^{-1}$. It was suggested that this behavior arises from strong spin fluctuations associated with the M-NM instability. As the effect of Sc substitution leads essentially to a decrease of the spacing between the Mn ions, increasing the M-NM instability, it became highly desirable to study the specific heat of YMn_2 under an applied hydrostatic pressure. The advantages of using an applied hydrostatic pressure are that the spacing between the Mn ions is decreased uniformly and can be varied continuously with no perturbation on the electronic structure.

2. Experiments and results

Polycrystalline samples of YMn_2 were prepared from starting Y and Mn elements of 99.9 % purity, under argon atmosphere, in an induction furnace allowing a quasi-levitation of the melt. After an annealing under vacuum (10^{-7} mbar) at 700°C during 72 hours, the obtained ingots were checked by the Debye-Scherrer X-ray method. No foreign phase was detected. The ingots were then cut by spark erosion into disc shapes of 6 mm of diameter and 2.5 mm of thickness. The sample was contained in a cell that was pressurized at room temperature at up to 12 kbar; the maximum pressure retained at low temperature, however, was 7.7 kbar. The pressure retained at low temperatures was measured by superconducting manometers at each end of the sample. The overall pressure gradient was of the order of 10% of the applied pressure. Since the magnetic ordering in YMn_2 is accompanied with a huge magnetovolume effect ($\Delta V/V \approx 5\%$) that disintegrates the sample into a coarse powder, care was taken to perform measurements at decreasing applied pressure from the available maximum value.

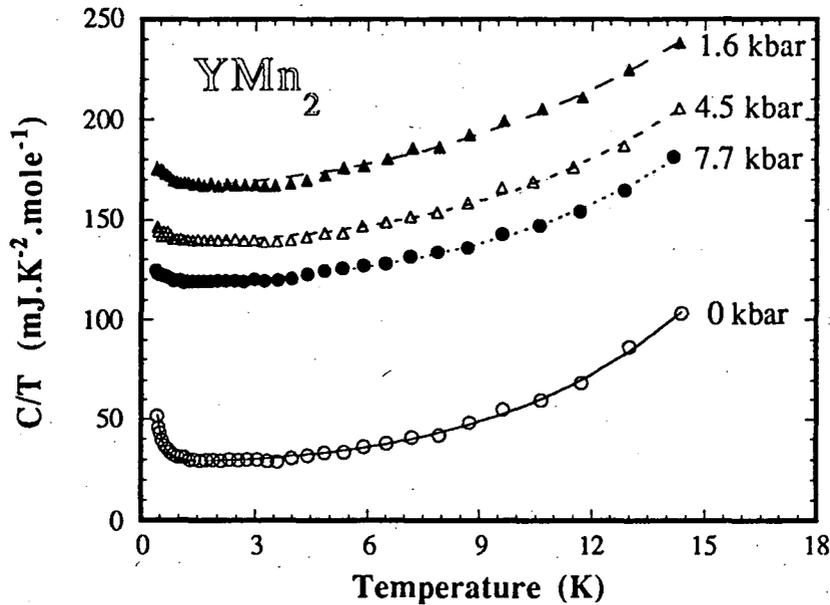


Fig.1 : C/T versus T at different applied pressures

Fig. 1 shows C/T vs T at different applied pressures. Except at 0 kbar where magnetic order is established, a large linear contribution to the specific heat is evident from large C/T values at low temperature. The experimental data were analyzed according to :

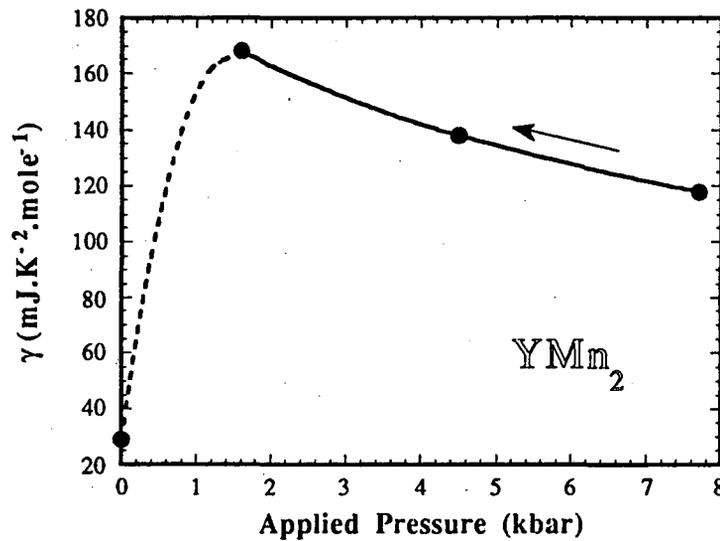
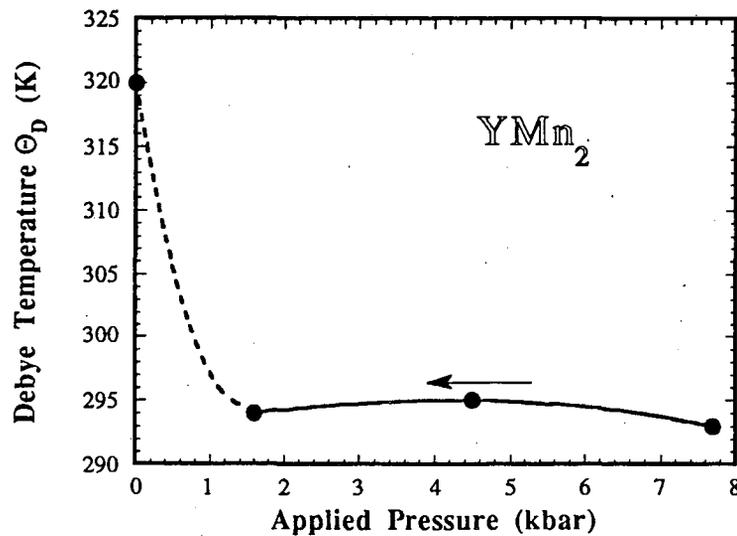
$$C = D_2 T^{-2} + \gamma T + B_3 T^3 + B_5 T^5$$

where the first term, corresponding to the upturn of the C/T versus T curves at very low temperature, reflects a hyperfine contribution; the second term expresses the electronic contribution and the third and fourth terms stand for the phonon contribution. We summarize in Table 1 the result of the fits. A large value of the linear contribution is observed at the applied pressures for which the ground state is paramagnetic. It increases when the applied pressure is

Table 1 : Pressure dependence of the coefficients of C

| Applied Pressure | D_2 mJ.K.mole^{-1} | γ $\text{mJ.K}^{-2}.\text{mole}^{-1}$ | B_3 $\text{mJ.K}^{-4}.\text{mole}^{-1}$ | B_5 $\mu\text{J.K}^{-6}.\text{mole}^{-1}$ |
|------------------|----------------------------------|-------------------------------------------------|----------------------------------------------|------------------------------------------------|
| 0 kbar | 1.75 | 29 | 0.177 | 0.895 |
| 1.6 kbar | 0.80 | 168 | 0.230 | 0.545 |
| 4.5 kbar | 0.55 | 138 | 0.227 | 0.433 |
| 7.7 kbar | 0.35 | 118 | 0.233 | 0.315 |

decreased from 7.7 kbar down to 1.6 kbar as shown in Fig.2. On the other hand, at zero applied pressure i.e. when the antiferromagnetic state is stabilized, the linear contribution is much lower.

Fig.2 : Pressure dependence of γ Fig.3 : Pressure dependence of the Debye temperature deduced from B_3

A Debye temperature, θ_D , has been deduced from the coefficient B_3 . Its variation with the applied pressure, shown in Fig.3, appears to be correlated with that of γ . It suggests that the crystal becomes less stiff as the amplitude of the fluctuations of the moments is enhanced.

3. Discussion

In general, enhancement of the linear contribution to the specific heat is expected in an itinerant electron paramagnet close to an antiferromagnet instability⁶. The measured values are very large and are associated with giant spin fluctuations easily excited even at low temperature⁷. The origin of these spin fluctuations is, however, not clear in detail. We suggest that their amplitude and their contribution to γ are strongly enhanced by the combined effects of the M-NM instability and the geometric frustration. It may be shown indeed, that a vanishing of moment may be induced by frustration⁸. Actually, there may be an analogy with heavy fermions since the two phenomena can be mapped in the same model⁹.

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