Magnetism of CoO under pressures of up to 7 GPa

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A first test experiment using a Paris-Edinburgh pressure cell has been carried out on the D20 diffractometer. A reinforcement with pressure of the antiferromagnetic order in cobalt monoxide has been observed. The pressure dependence of the ordered magnetic moment has also been established at room temperature up to 7.3 GPa.

The monoxides of *3d* transition metals, MnO, FeO, CoO and NiO form an interesting class of materials. Because of their apparently simple crystal and magnetic structures, they have been chosen as test materials for band theory models, and their electronic properties have been debated for a long time [1].

There exist several fundamental reasons to investigate these compounds under pressure.

On the one hand, theoretical calculations predict metallic behaviour for CoO and FeO at low pressures whereas they turn out to be insulators. This may be due to the neglect of the strong correlations between localized electrons. But, materials that are Mott or charge transfer insulators at low pressures are expected to become metallic with increasing pressure because both the *d* orbitals bandwidth and the screening increase. At high pressures, band theory calculations should thus be more reliable, but this has still to be experimentally confirmed.

On the other hand, previous studies [2,3,4,5], have shown that, in the *3d* transition metal monoxide series, orbitals are more compact in direct space than would be expected for a free transition metal ion. This feature could not result solely from an orbital contribution and the magnetization found on the oxygen is a good indication of the covalency effects present in these materials. Since the effect of pressure is to enhance exchange interactions, a better knowledge of the magnetic properties of these compounds under pressure is of prime interest.

Across the full series, FeO, MnO, CoO and NiO, phase magnetic collapse is predicted theoretically [6]. Because CoO is stoechiometric and has the lowest magnetic collapse transition, it should be the most amenable to experimental study. Up to now, only Bloch *et al.* [7] have measured the pressure dependence of the CoO magnetization up to 0.6 GPa. We have recently carried out neutron diffraction studies up to 7.3 GPa on D20 using the Paris-Edinburgh cells.

At ambient pressure, CoO orders at a Néel temperature T_N = 292 K in a type-II antiferromagnetic structure with a propagation vector τ = (1/2,1/2,1/2). At room temperature, i.e. in its paramagnetic phase, CoO crystallises in the NaCl structure (Fm3m, a = 4.261 Å).

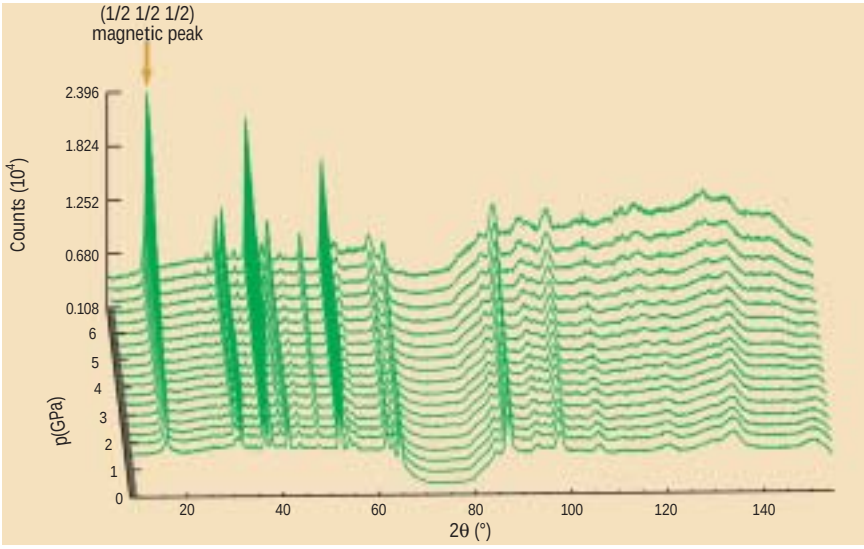

Figure 1: Pressure dependence of the CoO diffraction patterns measured at 300 K on the D20 diffractometer. The appearance and increase of the magnetic contribution is well evidenced with the (1/2,1/2,1/2) magnetic peak intensity variation. The sample volume is ~ 50 mm³, the accumulation time for each pattern is 40 min. The data have been normalized according to the strongest nuclear peak.

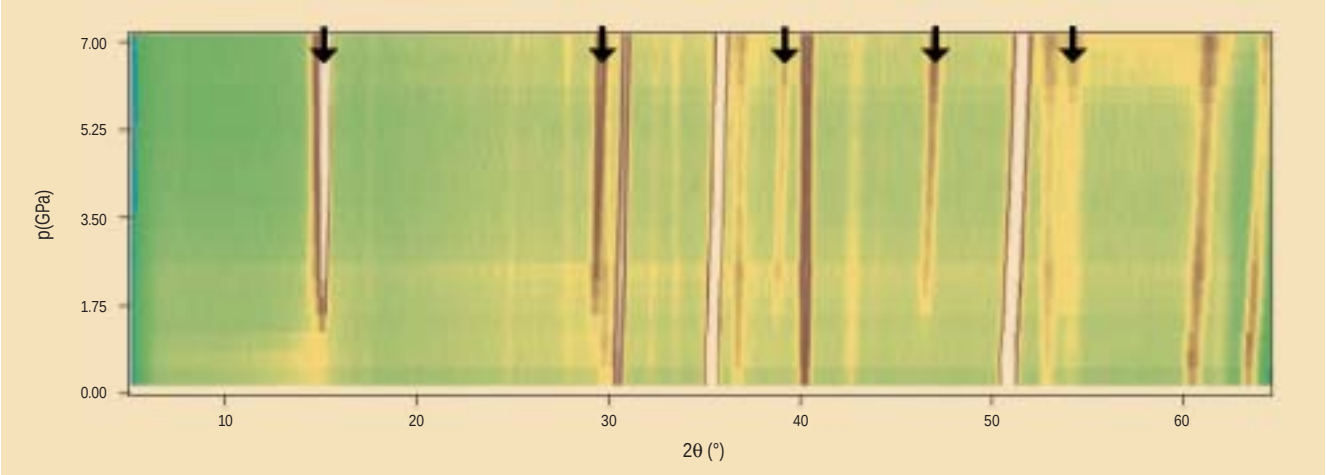

Figure 2: Low angular range zoom of the CoO diffraction pattern measured at 300 K under pressure. The magnetic contributions are marked with arrows.

The antiferromagnetic transition is accompanied by a cubic-to-monoclinic crystallographic distortion [8].

Figures 1 and 2 show the different normalized data patterns measured between p = 0 GPa and p = 7.3 GPa at 300 K. We clearly see the appearence of the magnetic signal at a pressure of ~ 1 – 1.5 GPa. Such an increase of T_N ($\Delta T_N \approx 8$ K) is consistent with the previous results of Bloch *et al.*, $\Delta(\log T_N)/\Delta(\log V) = -3.2$ [7]. The pressure dependence of the CoO magnetization at 300 K is depicted on figure 3. At 7.3 GPa, the saturation is still not achieved ($\mu_{sat} = 3.8\ \mu_B$).

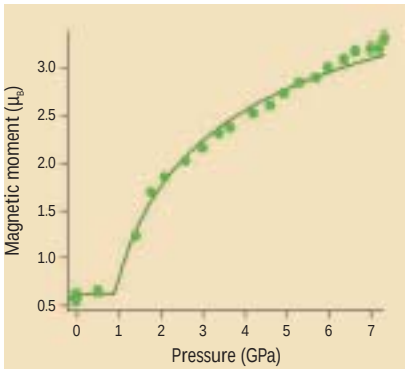

Figure 3: Pressure dependence of the CoO magnetization at 300 K, derived from Rietveld fits to the patterns shown in figure 2. The line is a guide for the eye.

These preliminary results on CoO under pressure are an important step towards a better understanding of the physics in the transition metal monoxides.

From a more general point of view, the pressure, like the temperature, is a fundamental parameter which governs the condensed matter properties. As it can be seen in the previous example of CoO, reaching high pressures is of prime interest for magnetic studies: indeed, slight structural distortions induced by pressure may have a large influence on the magnetic behaviour. In addition, pressure is also a unique tool for all the different structural aspects, since most of them result from a delicate balance between short- and long-range interactions which can be modified by applying a pressure.

The possibility of carrying out such experiments under a variable temperature would of course considerably increase the potentiality of this technique. Further work will also include the observation of lattice dynamics under high pressure.

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