

**Pressure-induced long-range magnetic ordering in cobalt oxide**Yang Ding,<sup>1,2,\*</sup> Yang Ren,<sup>3</sup> Paul Chow,<sup>1,2</sup> Jianzhong Zhang,<sup>4</sup> Sven C. Vogel,<sup>4</sup> Bjoern Winkler,<sup>5</sup> Jian Xu,<sup>1</sup> Yusheng Zhao,<sup>4</sup> and Ho-kwang Mao<sup>1,2</sup><sup>1</sup>HPCAT, Advanced Photon Source, Building 434E, Argonne National Laboratory, Argonne, Illinois 60439, USA<sup>2</sup>Geophysical Laboratory, Carnegie Institution of Washington, 5251 Broad Branch Road, Washington, DC 20015, USA<sup>3</sup>Experimental Facilities Division, Argonne National Laboratory, Argonne, Illinois 60439, USA<sup>4</sup>LANSCE, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA<sup>5</sup>Institut fuer Mineralogie, Abt. Kristallographie Universitaet Frankfurt Senckenberganlage, 30 D-60054 Frankfurt am Main, Germany

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A long-range magnetic ordering transition in cobalt oxide (CoO) was observed around 2.2 GPa at room temperature using neutron diffraction. The magnetic structure was type-II antiferromagnetic. High-resolution synchrotron x-ray diffraction showed no evidence of a tetragonal lattice distortion in CoO in the magnetically ordered phase. This result indicates that the tetragonal lattice distortion occurring with magnetic ordering in the low-temperature structure of CoO could be associated with cooperative Jahn-Teller effects (orbital ordering), which, however, are decoupled from magnetic ordering under high pressure.

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**I. INTRODUCTION**

The crystal, electronic, and magnetic structures of FeO and CoO, prototype narrow-band  $3d$  transition metal monoxides, under high pressure have long been of interest due to their complexities and challenges to theorists.<sup>1</sup> This paper discusses some of the fundamental problems in understanding these structures.

FeO and CoO are paramagnetic insulators with a rock salt structure above their Néel temperatures ( $T_N$ ), 198 K and 290 K, respectively. Below  $T_N$ , they transform into antiferromagnetic states. CoO distorts into a tetragonal symmetry accompanied by a small rhombohedral distortion,<sup>2</sup> whereas FeO distorts into a rhombohedral structure.<sup>3</sup> Upon compression at room temperature, FeO and CoO have been observed to distort into rhombohedral structures at  $\sim 15$  GPa (Ref. 4) and 43 GPa, respectively.<sup>5</sup> It has also been observed that  $T_N$  increases in CoO and FeO upon compression at room temperature.<sup>6,7</sup> Based on this linear pressure dependence of  $T_N$  (Refs. 6 and 7) and its similarity to the lattice distortion at low temperature, Yagi *et al.*<sup>4</sup> proposed in 1985 that the rhombohedral lattice distortion in FeO at  $\sim 15$  GPa should be associated with a magnetic ordering transition due to magnetic-elastic coupling. The conjecture of pressure-induced magnetic-elastic-driven lattice distortion in FeO at 15 GPa has been gradually accepted as correct.<sup>8,9</sup>

However, the conjecture faces the following problems: (1) after the lattice distortion occurred, no long-range magnetic ordering in FeO was observed at up to 20 GPa at room temperature;<sup>10</sup> (2) antiferromagnetic FeO at low temperature and high pressure was observed to exist in cubic but not rhombohedral symmetry,<sup>11</sup> implying that magnetoelastic coupling may not exist; (3) according to theoretical calculations, the observed lattice distortion in FeO is too large to be caused entirely by magnetoelastic coupling effects.<sup>12,13</sup>

On the other hand, since CoO has a similar pressure dependence of  $T_N$  and rhombohedral lattice distortion under pressure as FeO, it is natural to expect the occurrence of a pressure-induced magnetic-ordering-driven lattice distortion

in CoO, too. But the questions are: (4) why should CoO, with a higher  $T_N$ , distort under greater pressure than FeO, with a lower  $T_N$ ? and (5) if pressure just simply lifts  $T_N$ , why is no tetragonal lattice distortion observed in CoO under high pressure, as occurs at low temperature?

The above-mentioned questions about FeO and CoO imply that the pressure effects on their magnetic and crystal structures still are not fully understood. The complexity could arise from the interplay of spin, orbital, charge, and the lattice in these strongly correlated systems<sup>14</sup> under high pressure, demanding the combination of multiple techniques to reveal the change and the interaction of each individual degree of freedom. Therefore, in order to further investigate these problems, neutron diffraction and high-resolution synchrotron x-ray diffraction were combined with high-pressure techniques in this study to simultaneously investigate the effects of compression on both the magnetic and crystal structures of CoO. CoO not only has higher  $T_N$  than FeO (making it easier to magnetize under high pressure), but also does not have the defect-cluster problem that  $\text{Fe}_{1-x}\text{O}$  (Ref. 15) has. The results obtained from neutron diffraction clearly reveal a long-range magnetic ordering transition in CoO above 2.2 GPa at room temperature. However, no tetragonal lattice distortion is observed in the magnetically ordered phase.

**II. EXPERIMENTAL PROCEDURE**

CoO powder was purchased from Sigma-Aldrich with a purity of 99.99+ % (based on trace metal analysis). The high-pressure neutron experiments were performed at the HIPPO beamline of LANSCE, Los Alamos National Laboratory.<sup>16</sup> The sintered CoO powder sample was loaded into an amorphous  $\text{Zr}_2\text{P}_2\text{O}_7$  gasket and was then compressed at 300 K using a 500-ton toroidal anvil press TAP98 (a detailed description of TAP98 has been reported elsewhere<sup>17</sup>). Neither a pressure standard nor a pressure medium was used in the sample chamber in order to obtain clean diffraction patterns, and the pressure was determined from the isothermal equation of the state of CoO ( $K_0=180$  GPa and  $K'_0=3.82$ ).<sup>5</sup> The

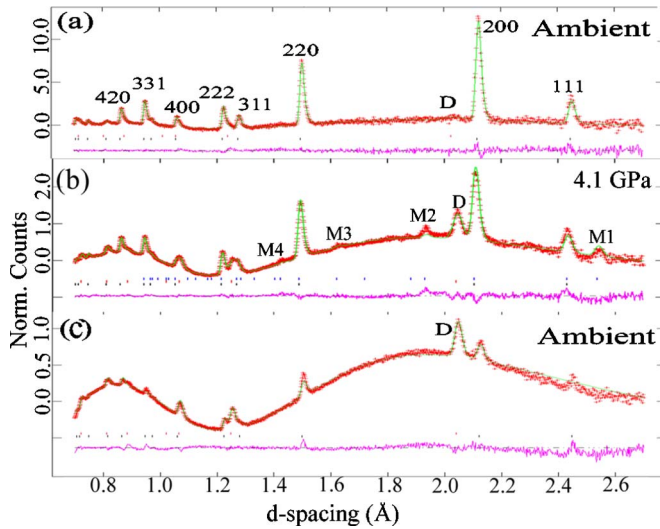


FIG. 1. (Color online) Neutron diffraction patterns of CoO collected at (a) ambient pressure (lattice parameter  $a=4.261$  Å), (b) 4.1 GPa (lattice parameter  $a=4.230$  Å), and (c) ambient pressure (released from 9.0 GPa). The horizontal axis of the plot is  $d$  spacing in Å and the vertical axis is the normalized counts per second.  $M$ : magnetic peak;  $D$ : diamond peak.

neutron diffraction data were refined using the general structure analysis system (GSAS) program.<sup>18</sup> As the angular resolution of neutron diffraction at HIPPO is insufficient to discern the possible subtle lattice distortion of CoO, high-resolution synchrotron x-ray experiments were performed on CoO, which was hydrostatically compressed in a diamond anvil cell at sector 11 IDC, Advanced Photon Source, Argonne National Laboratory. With monochromatic x-ray photons of 115.125 KeV, a Si (220) analyzer, and a germanium solid-state point detector, the diffraction angular resolution ( $\Delta\theta/2\theta$ ) was determined to be  $<0.05\%$ . The sintered CoO powdered chip was loaded into a symmetric diamond anvil cell, together with a ruby pressure marker and pressure medium, a mixture of methanol (80%) and ethanol (20%). The pressure was determined from the fluorescence peak shift of a ruby single crystal.<sup>19</sup>

### III. RESULTS

Figure 1 shows high-pressure neutron diffraction patterns collected at ambient pressure, 4.1 GPa, and on the sample recovered after the pressure was released. Figure 2 emphasizes the relative intensity changes of magnetic peak  $M2$  and nuclear peak 200, as a function of pressure. Above 2.6 GPa, the magnetic peaks  $M1$ ,  $M2$ ,  $M3$ , and  $M4$  are first observed, indicating the occurrence of a long-range magnetic ordering transition at room temperature. Such a transition was corroborated by a continuous increase in the relative intensity ratio of the magnetic peaks to the nuclear peak 200 with increasing pressure. After the pressure was released to ambient conditions, all magnetic peaks disappeared, indicating that the magnetic ordering transition is reversible. By fitting the intensity ratio of magnetic peak  $M2$  and peak 200 as a function of pressure, the paramagnetic-antiferromagnetic or-

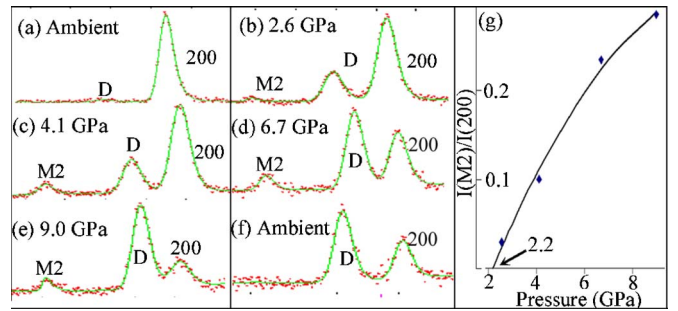


FIG. 2. (Color online) The enlarged diffraction patterns consisting of magnetic peak  $M2$  ( $3/2, 3/2, 1/2$ ) and nuclei peak 200 at (a) ambient pressure, (b) 2.6 GPa, (c) 4.1 GPa, (d) 6.7 GPa, (e) 9.0 GPa, and (f) ambient pressure (released from 9.0 GPa).  $M$ : magnetic peak;  $D$ : diamond peak. (g) The variation of the intensity ratio,  $I_{M2}/I_{200}$ , with pressure. The magnetic transition pressure ( $\sim 2.2$  GPa) is obtained from fitting the data of  $I_{M2}/I_{200}$  up to 9.0 GPa.

dering transition pressure was determined to be 2.2 GPa at room temperature, as shown in Fig. 2(g). As a result, the  $T_N$  rate of increase with pressure was determined to be 4.5 K/GPa, which is comparable with previous results.<sup>20</sup> The structure refinement performed with GSAS, shown in Fig. 1, includes phases of CoO ( $Fm-3m$ ) with AF-II magnetic structure, diamond ( $Fd-3m$ ), and amorphous  $ZrP_2O_7$ . The results indicate that CoO has cubic symmetry within the investigated pressure range, and the  $d$  spacing, as well as intensity of magnetic peaks, match well with those derived from the AF-II-type model using a spin propagation vector  $\tau=[1/2, 1/2, 1/2]$ .<sup>23</sup> Consequently, the magnetic peaks  $M1$ ,  $M2$ ,  $M3$ , and  $M4$  can be indexed as  $3/2, 1/2, 1/2, 3/2, 3/2, 1/2, 5/2, 1/2, 1/2, 5/2, 3/2, 1/2$ , respectively. No lattice distortion could be discerned within the neutron diffraction angular resolution of  $\Delta d/d \sim 0.1\%$  (determined as full width at half-maximum (FWHM)<sub>peak</sub>/ $d_{peak}$ ).

Figure 3(a) displays the results from the high-resolution x-ray diffraction experiments on CoO. The data were collected from synchrotron radiation at ambient pressure, 1.3 GPa, 3.5 GPa, 6.7 GPa, and 10.3 GPa. The diffraction data at ambient pressure were collected from loose powder, and the FWHM of 111 and 200 are  $0.0027^\circ$  and  $0.0047^\circ$ ,

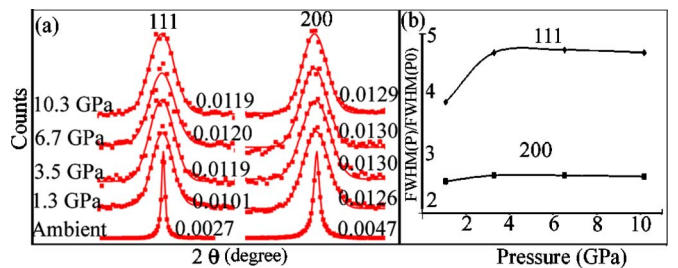


FIG. 3. (Color online) (a) The 111 and 200 diffraction peaks of CoO collected with synchrotron x rays of energy 115.125 KeV under high pressure. The numbers on the right side of the diffraction peaks are the corresponding FWHMs. (b) The variation of normalized FWHM as a function of pressure for the 111 and 200 peaks.  $P_0$  refers to ambient pressure.

respectively, corresponding to  $2\theta$  values of  $2.509^\circ$  and  $2.897^\circ$ . Once the sample was sintered into a dense plate, loaded into the diamond anvil cell, and compressed to 1.3 GPa, the FWHM of the 111, 200, 220 peaks broaden to three to four times larger than those at ambient pressure, indicating that strain broadening was introduced by sintering. However, the FWHM for the 200 peak stayed unchanged from 1.3 GPa to 10.3 GPa, as shown in Fig. 3(b). Based on a  $T_N$  increasing rate of 4.5 K/GPa and a  $T_N/T$  ratio as 1.12, the diffraction pattern at 10.3 GPa and room temperature is equivalent to that at 258 K (or perhaps even lower, since  $T_N$  increases faster under hydrostatic compression than under nonhydrostatic compression<sup>21</sup>) and ambient pressure. According to high-resolution x-ray diffraction results at low temperature,<sup>2</sup> the tetragonal lattice distortion  $(a-c)/c$  of CoO at 258 K is about 0.45%, corresponding to a  $0.003^\circ$  increase in FWHM of peak 200 at an x-ray energy of 115.125 KeV. However, neither splitting nor broadening of the 200 peak is seen in Figs. 3. In conjunction with previous high-pressure experiments of CoO up to 104 GPa, during which a tetragonal lattice distortion was not observed,<sup>2</sup> it is safe to conclude that under static compression at room temperature there is no tetragonal lattice distortion associated with the magnetic ordering transition. However, compared with the 200 peak, the larger broadening of the 111 peak after sintering and the more rapid increase of its FWHM with pressure immediately after magnetic transition could indicate softening of the elastic shear constant  $C_{44}$ , and hence a rhombohedral lattice distortion. Though the rhombohedral lattice distortion is so small [at 93 K, the shear strain for the rhombohedral is only about  $10^{-5}$  (Ref. 2)] that the splitting of the 111 is not resolvable within the low-pressure range, the compression of CoO up to 104 GPa confirmed the rhombohedral lattice distortion around 43 GPa.<sup>5</sup> An overestimated structural transition pressure in CoO is likely resulted from the low diffraction resolution of the energy dispersive diffraction method.<sup>22</sup>

#### IV. DISCUSSION

Pressure-induced long-range magnetic ordering observed in CoO at  $\sim 2$  GPa, but not in  $\text{Fe}_{1-x}\text{O}$  up to 20 GPa, suggests that pressure indeed can increase  $T_N$  of CoO (or FeO), but the relationship between  $T_N$  and pressure is not linear as previous understanding.<sup>6,7</sup> Hence, the absence of long-range magnetic ordering to up 20 GPa at room temperature in  $\text{Fe}_{1-x}\text{O}$  was likely because the pressure was still not high enough. The reason for the magnetic transition in CoO (or  $\text{Fe}_{1-x}\text{O}$ ) is because the pressure-induced volume reduction increases  $T_N$ .<sup>23,24</sup> Once  $T_N$  reaches room temperature, i.e.,  $T_N/T > 1$ , magnetic ordering occurs. However, the pressure-induced increase in  $T_N$  may be sustained in only a relatively low-pressure regime, since magnetic moments tend to decrease and eventually disappear as pressure is increased. This can be understood in the Stoner model,<sup>25</sup> in the sense that increasing pressure generally leads to an increase in bandwidths and a decrease of effective density of states, whereas the effective Stoner parameter is insensitive to pressure variation. This also happens in the Hubbard model, since the parameters of the two models are closely related. The mag-

netism collapse in CoO GPa (Ref. 26) indicates the pressure effect on magnetism is not monotonic and hence the  $T_N$  rate of change with pressure cannot be linear at relatively high pressure. It can be inferred that  $T_N$  increases with pressure to a certain value and then starts to decrease. Due to this, the pressure required to magnetize materials with a lower  $T_N$  (i.e., FeO with  $T_N=198$  K) may be higher than that expected from the rate obtained at low pressure. This could be the reason why the magnetic structure was not observed in FeO up to 20 GPa at room temperature.<sup>10</sup>

It is known that, with AF-II ordering, MnO, FeO, and NiO are distorted trigonally along the [111] direction, whereas CoO has a large tetragonal distortion below its  $T_N$ .<sup>3</sup> It was recently realized that the low-temperature structure has monoclinic symmetry, consisting of a small trigonal distortion accompanying the much larger tetragonal distortion.<sup>2</sup> A single-crystal neutron diffraction study revealed a weak type-I AF ordering, which is related to the tetragonal distortion.<sup>27</sup> Though much effort has been made, both experimentally and theoretically, to understand the origin of the tetragonal lattice distortion below  $T_N=290$  K in CoO, the situation remains unclear. However, the lack of a tetragonal lattice distortion under high pressure in CoO after magnetic ordering found in this study indicates that a tetragonal lattice distortion in the low-temperature structure accompanying magnetic ordering should be associated with a cooperative Jahn-Teller distortion (orbital ordering), but it is decoupled with magnetic ordering under high pressure.

According to Kanamori,<sup>12,13</sup> the observed tetragonal distortion at low temperature is two orders of magnitude larger than his calculations based on the normal magnetostriction. Thus, the giant tetragonal distortion in the low-temperature structure of CoO has to be induced predominantly by a cooperative Jahn-Teller effect (orbital ordering),<sup>10,11,28</sup> while the small trigonal lattice distortion is caused by magnetostriction. CoO has  $\text{Co}^{2+}$  ions with the  $3d^7$  configuration surrounded by six oxygen atoms. In the cubic crystal field, the low-energy  $t_{2g}$  levels occupied by five electrons have a three-fold orbital degeneracy, thus making  $\text{Co}^{2+}$  ions Jahn-Teller active. Following the theories of Kugel and Khomskii,<sup>28</sup> the orbital degeneracy can be lifted by either spin-orbital coupling or orbital ordering, which give different types and magnitudes of lattice distortion and magnetic anisotropy. Kugel and Khomskii (Ref. 28) explained the lattice property and magnetic structure in terms of orbital ordering, which is clearly evidenced by the anisotropic electron densities of  $\text{Co}^{2+}$  ions at low temperature obtained by a  $\gamma$ -ray diffraction.<sup>29</sup> Both the type of tetragonal lattice distortion and magnetic anisotropy in CoO support the orbital-ordering picture. Jauch *et al.*,<sup>29</sup> however, raised doubts about the Jahn-Teller effect, with a particular concern about the simultaneous occurrence of the tetragonal distortion and the magnetic ordering. While orbital ordering and magnetic ordering often occur at different temperatures in  $e_g$  systems,<sup>1</sup> it is not uncommon for  $t_{2g}$  systems to undergo orbital ordering in the vicinity of (such as in  $\text{LaVO}_3$  and other compounds<sup>29,30</sup>) or precisely at [such as in  $\text{LaTiO}_3$  (Ref. 31) and  $\text{YVO}_3$  (Ref. 32)] the magnetic transition temperature. The nature of the exchange interaction in a magnetic material containing Jahn-Teller ions depends strongly on the spin cor-



relation function, which varies with the temperature and is dependent on details of orbital ordering. Therefore, it is possible that, above a certain temperature, the cooperative Jahn-Teller effect (or orbital ordering) does not occur. As in CoO, where the larger Jahn-Teller tetragonal lattice distortion is decoupled with magnetic ordering at high-pressure room temperature, this kind of decoupling could also occur in other similar materials, i.e., FeO. Indeed, the giant rhombohedral lattice distortion was observed to be decoupled with magnetic ordering,<sup>11</sup> indicating that the rhombohedral lattice distortion observed in FeO at 15–18 GPa and room temperature is not necessarily driven by magnetic ordering, but is likely caused by other mechanisms.

## V. CONCLUSIONS

In summary, we have observed a reversible long-range magnetic ordering transition in CoO using neutron diffraction at around 2.2 GPa and room temperature. The magnetic structure is type-II antiferromagnetic with spin wave vector  $\tau=[1/2, 1/2, 1/2]$ . No tetragonal lattice distortion is discerned with high-resolution synchrotron x-ray diffraction on hydrostatically compressed CoO. The lack of a tetragonal lattice distortion under high pressure in CoO indicates that the orbital ordering is decoupled from magnetic ordering under high pressure, though they occur simultaneously at low temperature. The results also shed light on problems in FeO

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- <sup>23</sup> $T_N$  is associated with the exchange interaction through the relation  $2JzS(S+1)/3k$ , (where  $J$  is the exchange interaction between next-nearest neighbors,  $z$  is the total number of nearest and next-nearest neighbors,  $S$  is the spin vector, and  $k$  is the Boltzmann constant) and  $J$  is proportional to  $a^{-10}$  ( $a$ : unit cell parameter) (Ref. 24). Therefore, compression causes volume reduction to result in the increasing of  $T_N$ . For CoO, the next-nearest-neighbor ( $180^\circ$ ) antiferromagnetic exchange  $J$  is much larger than the nearest-neighbor ( $90^\circ$ ) ferromagnetic coupling, so that the latter can be ignored.
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