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Low-energy nuclear spin excitations in CoO

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We investigated the low-energy excitations in CoO in the μeV range by a back-scattering-neutron spectrometer. The energy scans on a CoO single crystal revealed inelastic peaks at $E=2.05\pm 0.02$ μeV at $T=3.5$ K on both energy-gain and energy-loss sides. The inelastic peaks move gradually toward lower energy with increasing temperature and finally merge with the elastic peak at the electronic magnetic ordering temperature $T_N \approx 290$ K. We interpret the inelastic peaks to be due to the transition between hyperfine-split nuclear level of the ^{59}Co isotopes with spin $I=7/2$.

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The statics and dynamics of ordered nuclei in magnetic crystals have attracted some attention from the condensed-matter physicists. The experiments however are difficult due to the very low temperatures needed and also the due to the required energy resolution of the order of a μeV or better of the neutron spectrometers to be used for such studies. Blume and Schermer[1] were the first to show the potential application of the neutron-scattering technique to study nuclear spin systems and calculated the relevant differential neutron-scattering cross sections and final-state polarization for elastic-scattering processes. Later Word *et al.*[2] extended these calculations to inelastic-scattering processes. Heidemann *et al.*[3–9] investigated the hyperfine fields in Co- and V-based compounds by using high-resolution back-scattering-neutron spectrometer. The hyperfine splitting lies typically in the energy range of a few μeV. The inelastic spin-flip scattering of neutrons from the nuclear spins can yield this information provided the neutron spectrometer has the required resolution of about 1 μeV or less and also the incoherent scattering of the nucleus is strong enough. It was established that the hyperfine field produced at the nucleus is roughly but not exactly proportional to the electronic magnetic moment of the $3d$ shell. Chatterji *et al.*[10–16] and also Przenioslo *et al.*[17] investigated low-energy nuclear excitation in Nd and Nd-based compounds. Recently Ehlers *et al.*[18] reported observation of nuclear spin excitations in pyrochlore structure spin ice compound $Ho_2Ti_2O_7$.

The method of determining the hyperfine splitting of the nuclear levels by spin-flip scattering of neutrons is now well established.[3] The relevant neutron-scattering process can be summarized as follows: If neutrons with spin $\mathbf{s}$ are scattered from nuclei with spins $\mathbf{I}$, the probability that their spins will be flipped is 2/3. The nucleus at which the neutron is scattered with a spin flip, changes its magnetic quantum number M to $M\pm 1$ due to the conservation of the angular momentum. If the nuclear ground state is split up into different energy levels E_M due to the hyperfine magnetic field or an electric quadrupole interaction, then the neutron spin flip produces a change in the ground-state energy $\Delta E=E_M-E_{M\pm 1}$. This energy change is transferred to the scattered neutron. If there is only one isotope then one expects a central elastic peak and two inelastic peaks of approximately equal intensities. The element Co is such a case because the isotope ^{59}Co has 100% natural abundance and therefore the isotope incoherent-scattering cross section is zero. Cobalt has the natural abundances of 100% for the ^{59}Co isotopes with nuclear spin $I=7/2$ and its incoherent-scattering cross section[19] is relatively large, 4.8 ± 0.3 b. Therefore Co and Co-based compounds are suitable for the studies of nuclear spin excitations. In fact Heidemann *et al.*[6,7] studied nuclear spin excitations in ferromagnetic Co and Co-P amorphous alloys and also Co-based intermetallic compounds $LaCo_{13}$, $LaCo_5$, YCo_5, and $ThCo_5$. During the present investigation we studied low-energy nuclear spin excitations in antiferromagnetic transition-metal oxide CoO.

CoO belongs to the family of transition-metal oxides, which have been the subject of intensive investigations. The transition-metal oxides with narrow d bands from strongly correlated electron systems or a Mott-Hubbard system in which the Coulomb interactions between electrons lead to breakdown of the conventional band theory. CoO along with other transition-metal oxides MnO, FeO, and NiO crystallize with the face-centered-cubic NaCl-type structure in the $Fm\bar{3}m$ space group. CoO orders[20–27] below $T_N \approx 290$ K with the type-II structure with the propagation vector $\mathbf{k}=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$. Ferromagnetic (111) are antiferromagnetically stacked along the [111] direction.

We performed inelastic neutron-scattering experiments on a CoO single crystal by using the high-resolution back-scattering-neutron spectrometer SPHERES of the Jülich Centre for Neutron Science located at the FRMII reactor in Munich. The wavelength of the incident neutrons was $\lambda=6.271$ Å. A large CoO single of conical shape with a diameter of about 5–15 mm and length of about 25 mm was fixed on the cold tip of a Displex refrigerator with its [100] crystallographic direction vertical. We observed inelastic signals in CoO at energies $E=2.05\pm 0.02$ μeV on both energy gain and loss sides at $T=3.5$ K. The energy of the inelastic signal decreases continuously as the temperature is increased and finally merges with central elastic peak at $T_N \approx 290$ K. Figure 1 shows typical energy spectra of CoO at several temperatures. The spectra are the result of summing up the counts of the individual detectors placed at different scattering angles. The inelastic signals have resolution limited widths at least at low temperatures. The inelastic peaks move toward the central elastic peak as the temperature is increased and at the ordering temperature they just merge with

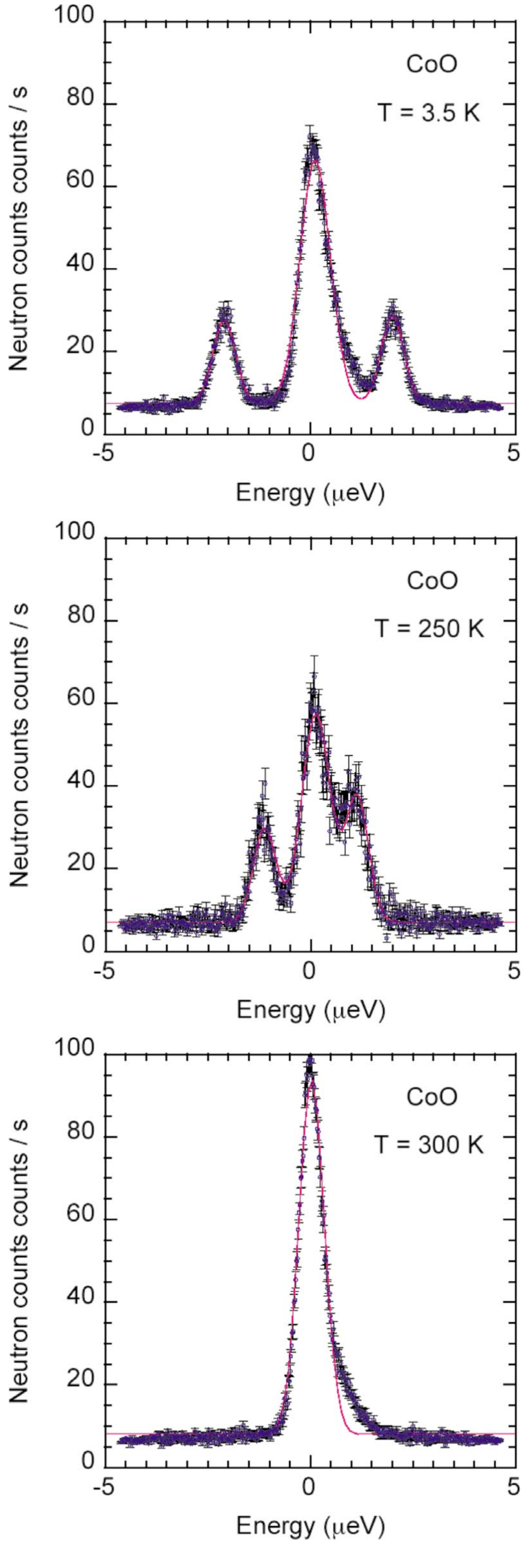

FIG. 1. (Color online) Typical energy spectra of CoO at several temperatures.

the central elastic peak thereby causing an increase in intensity of the elastic peak. We fitted the two inelastic and the central elastic peaks with Gaussian functions. The shape of the elastic peak at $E=0$ at low temperature is essentially determined by the resolution function of the back-scattering spectrometer. The resolution function was found to be asymmetric with a shoulder on the positive-energy side. So the Gaussian function can describe the resolution function only approximately. We attribute the asymmetric shape due to the deviation from the perfect back-scattering geometrical situation. The asymmetric line shape hindered us to get a good determination of the position, intensity, and width of the inelastic peaks especially the one at the positive-energy side close to the ordering temperature at which the inelastic peak are very close to the central elastic peak. We interpret the inelastic signal observed in CoO due to the excitations of the ^{59}Co nuclear spins $I=\frac{7}{2}$. In a first approximation one can consider these inelastic peaks to arise due to the transitions between the hyperfine-field-split nuclear levels.

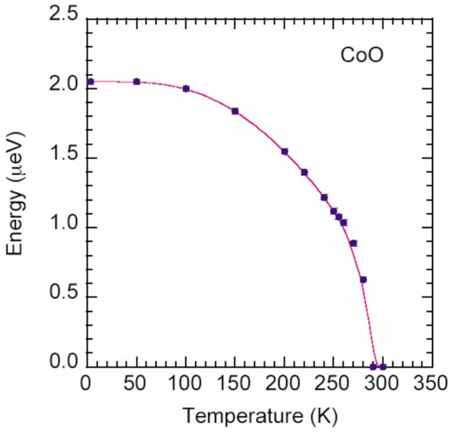

FIG. 2. (Color online) Temperature variation in the energy of the inelastic peak of CoO. The continuous line is just a guide to the eye.

Figure 2 shows the temperature variation in the energy of the inelastic peak of CoO. The energy of the inelastic peak can be considered to be the order parameter of the phase transition. Due to the limited neutron beam time we could measure enough data close to the antiferromagnetic phase-transition temperature and therefore the critical exponent β and the Néel temperature T_N could not be determined accurately from the present experiment. However the data are consistent with the reported[27] Néel temperature $T_N \approx 290$ K. The identification of the order parameter with the energy of the inelastic signal works well in a single magnetic sublattice system such as CoO. It has been shown more rigorously in our recent paper[16] on $NdAl_2$ that the identification of the order parameter with the energy of the inelastic signal or the hyperfine splitting of the nuclear levels is justified. Our previous study[15] on $NdMnO_3$ showed that such identification of the order parameter with the energy of the inelastic peak fails for the more complex systems with two magnetic sublattices.

The intensity of the inelastic peak at $T=3.5$ K is about one third of that of the elastic peak. Natural Co has only one isotope and therefore gives no isotope incoherent scattering. The incoherent-scattering cross section of O is zero and therefore do not contribute to the intensity of the incoherent elastic peak. The remaining possibilities are the contributions from the sample holder and coherent Bragg peaks. The sample holder consists of Al which has a very small incoherent-scattering cross section of 0.0092 ± 0.0007 b. To decrease the background we used Cd which has a large

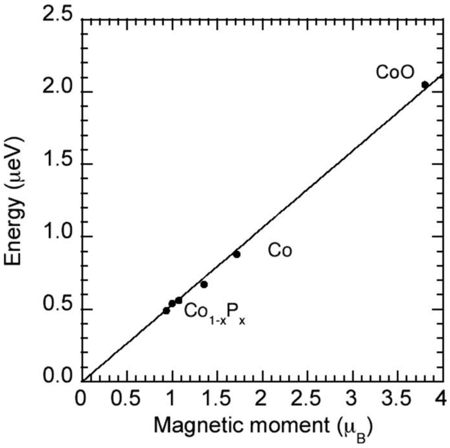

FIG. 3. Plot of the energy of inelastic signal vs ordered electronic moment of Co-based materials.

incoherent-scattering cross section of 2.4 ± 0.7 *b*. These are the possible origin of the excess intensity in the elastic peak. Heidemann *et al.*[4,5,8] observed similar excess intensity at the elastic peak in several experiments on vanadium oxides.

Table I gives the ordered electronic moment of CoO and the energy of Co nuclear spin excitations of CoO determined during the present investigations along with the similar data obtained by Heidemann *et al.*[6] in Co and Co-P amorphous alloys. Figure 3 shows a plot of energy of inelastic peaks observed in CoO, Co, and Co-P alloys[6] vs the corresponding saturated electronic magnetic moment of Co in these compounds. The data lie approximately on a straight line showing that energy of inelastic peak or the hyperfine splitting of the nuclear level is approximately proportional to the electronic magnetic moment. The slope of the linear fit of the data gives a value of 0.531 ± 0.006 $\mu eV/\mu_B$. It is surprising that this linear relationship is valid for metallic Co and Co-P alloys as well as for insulating CoO. However similar linear relationship has been obtained for metallic Nd and Nd-based several metallic and insulating compounds.[12–16]

TABLE I. Ordered electronic moment of Co and the energy of Co nuclear spin excitations.

Compound	Moment (μ_B)	ΔE (μeV)	Reference
CoO	3.80(6)	2.05(1)	Present work
Co	1.71	0.88	4
$Co_{0.873}P_{0.127}$	1.35	0.67	4
$Co_{0.837}P_{0.161}$	1.0	0.54	4
$Co_{0.827}P_{0.173}$	1.07	0.56	4
$Co_{0.82}P_{0.18}$	0.93	0.49	4

In conclusion we have investigated the low-energy excitations in CoO by inelastic neutron scattering with a backscattering-neutron spectrometer. The present results together with the results on Co and Co-P amorphous alloys studied by Heidemann *et al.*[6,7] have shown that the ordered electronic magnetic moment of Co is linearly proportional to the energy of excitations.

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