ISSN 1063-7834, Physics of the Solid State, 2008, Vol. 50, No. 9, pp. 1723–1726. © Pleiades Publishing, Ltd., 2008.

PROCEEDINGS OF THE XIII FEOFILOV SYMPOSIUM "SPECTROSCOPY OF CRYSTALS DOPED BY RARE-EARTH AND TRANSITION-METAL IONS"

(Irkutsk, July 9–13, 2007)

Magnetic Ordering in $Co_cMg_{1-c}O$ Solid Solutions[1]

N. Mironova-Ulmane[a], U. Ulmanis[a], A. Kuzmin[a], I. Sildos[b], M. Pärs[b], M. Cestelli Guidi[c], M. Piccinini[c,d], and A. Marcelli[c]

[a] *Institute of Solid State Physics, University of Latvia, Riga, LV-1063 Latvia*
[b] *Institute of Physics, University of Tartu, Tartu, EE-2400 Estonia*
[c] *Laboratory Nazionali di Frascati, Istituto Nazionale di Fisica Nucleare, Frascati, Italy*
[d] *Dipartimento di Scienze Geologiche, Universitá Roma Tre, Roma, Italy*

Abstract—The influence of dilution by diamagnetic ions on the magnetic ordering in single-crystal $Co_cMg_{1-c}O$ solid solutions was studied by Raman spectroscopy and magneto-optical microscopy in a wide range of temperatures ($6 < T < 200$ K). Far infrared absorption measurements of antiferromagnetic resonance (AFMR) were also performed for pure CoO. It was found that the domain structure and the contribution from Brillouin zone center magnons to Raman scattering and AFMR disappear well below the Néel temperature, whose value was determined from neutron diffraction and magnetic susceptibility measurements.

PACS numbers: 75.25.+z, 75.50.Ee, 78.30.-j

DOI: 10.1134/S1063783408090266

1. INTRODUCTION

The $Co_cMg_{1-c}O$ system is a nice example of diluted antiferromagnets [1, 2]. It forms a continuous series of solid solutions, whose magnetic properties vary with composition from antiferromagnetic-like behavior with a Néel temperature $T_N = 290$ K for pure CoO to diamagnetic-like for pure MgO. For intermediate compositions with $c > 0.5$, the paramagnet-to-antiferromagnet phase transition occurs upon cooling. Note that paramagnetic CoO has a NaCl-type crystal structure (space group $Fm3m$) and that the antiferromagnetic ordering in it is associated with a cubic-to-monoclinic transition (space group $C2/m$ with two CoO units per cell) [3].

In this paper, we report the results of Raman spectroscopy studies and microscopic magneto-optical observations of domain structure in single-crystal $Co_cMg_{1-c}O$ solid solutions performed in a wide temperature range ($6 < T < 200$ K). Far infrared (IR) measurements of antiferromagnetic resonance (AFMR) and magnetic Raman scattering in pure single-crystal CoO are used for comparison.

2. EXPERIMENTAL

The $Co_cMg_{1-c}O$ samples used in the present work were single crystals obtained from polycrystalline solid solutions through chemical transport reaction (the "sandwich" technique) on the (100) face of MgO single crystals. Polycrystalline $Co_cMg_{1-c}O$ solid solutions were prepared from appropriate amounts of aqueous solutions of $Mg(NO_3)_2 \cdot 6H_2O$ and $Co(NO_3)_2 \cdot 6H_2O$ salts, which were mixed and slowly evaporated. The remaining dry "flakes" were heated to 500–600°C to remove NO_2 completely. The obtained polycrystalline solid solutions were pressed and annealed at 1200°C in air for 100 h and then rapidly cooled to room temperature.

Far IR optical absorption measurements were performed at the SINBAD beamline of the DA FNE synchrotron radiation source (Frascati, Italy) using a Bruker Equinox 55 interferometer modified to work in vacuum in the far IR range with a liquid He-cooled bolometer. Temperature-dependent measurements were done for single-crystal CoO(100)/MgO(100) in the range from 100 to 300 cm^{-1} using a liquid-helium cryostat in the temperature range from 5 to 200 K.

Low-temperature optical microscopy and Raman studies were carried out by using a home-made micro-Raman spectrometer based on a standard Olympus microscope, equipped with a single-grating Andor Shamrock 303i spectrometer (focal length 303 mm, 600-lines/mm grating with a resolution of about 10 cm^{-1}) and a TE cooled Andor Newton EMCCD camera. The Nd–YAG 532 nm laser excitation was used with an intensity of about 5 mW on illuminated area. A 50× objective was used, so that sampling volume was a cylinder with a diameter of 1.5 μm and a depth of about 14 μm. To carry out low-temperature measurements, a special Utreks-type cold-finger helium cryostat was designed.

[1] The text was submitted by the authors in English.

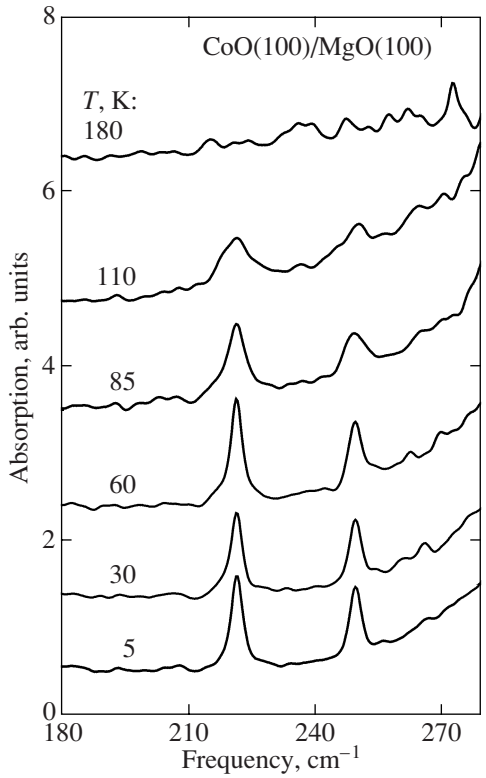

Fig. 1. Far IR optical absorption spectra of single-crystal CoO(100)/MgO(100). Two AFMR bands at 221 and 249 cm^{-1} are well visible below 180 K.

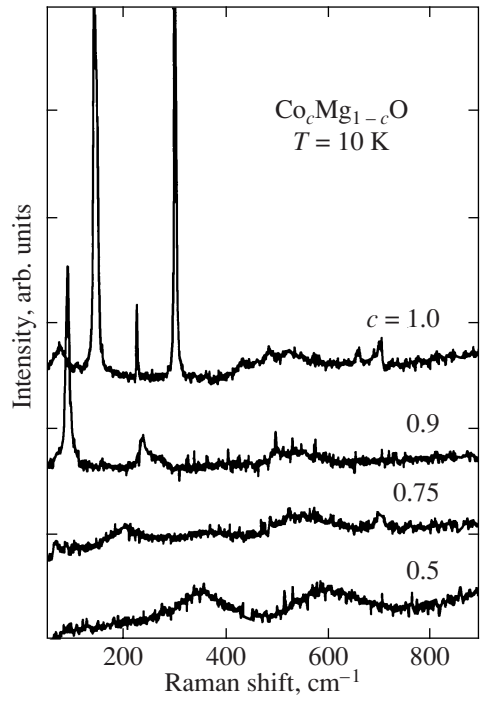

Fig. 2. Raman spectra of $Co_cMg_{1-c}O$ solid solutions at T = 10 K. Two sharp peaks due to magnon excitations are well visible for c = 1.0 and 0.9.

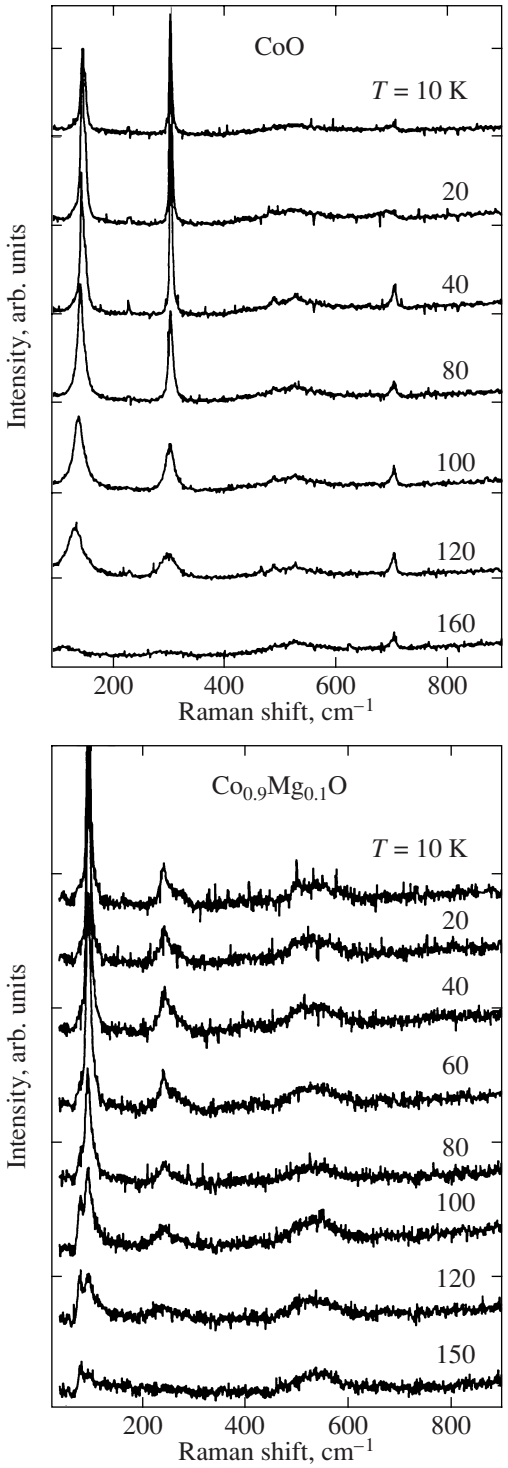

Fig. 3. Temperature dependence of Raman scattering in CoO and $Co_{0.9}Mg_{0.1}O$ solid solutions. The small peak at 700 cm^{-1} for CoO is due to an admixture of the Co_3O_4 phase.

3. RESULTS AND DISCUSSION

The far IR spectra for single-crystal CoO are shown in Fig. 1. The two AFMR bands at 221 and 249 cm^{-1} were reliably measured below 180 K and are in good

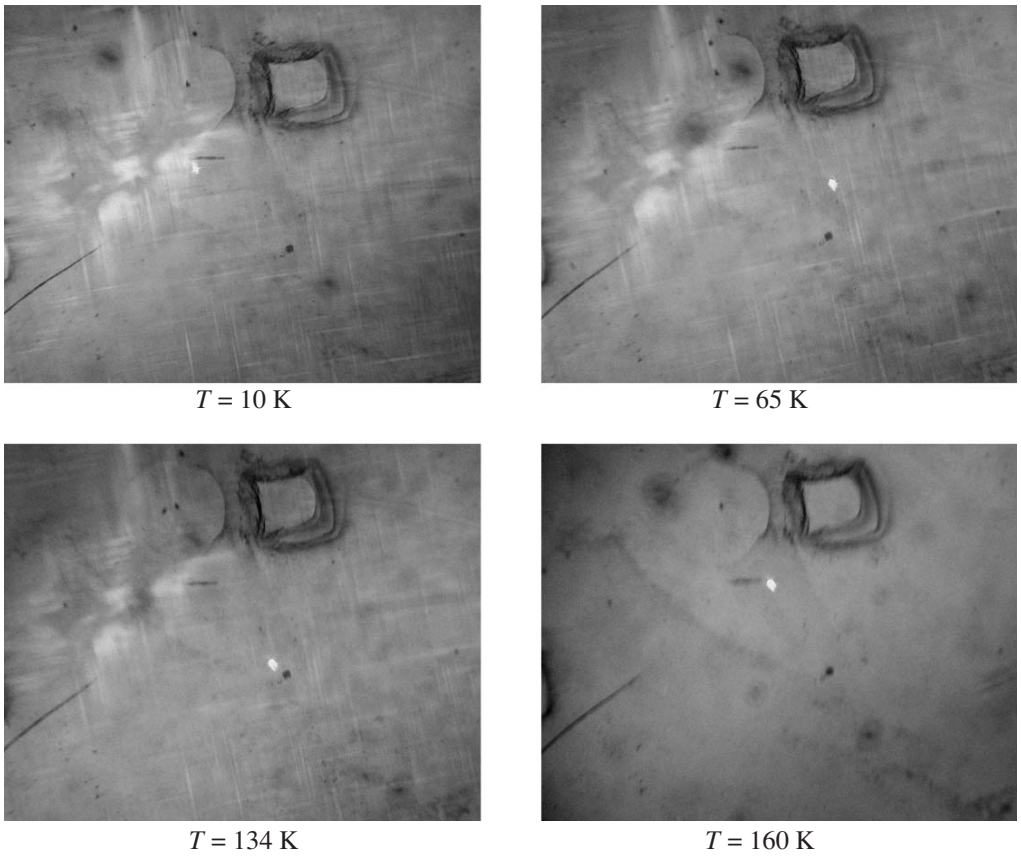

Fig. 4. Twin domain structure of single-crystal $Co_{0.9}Mg_{0.1}O$ observed in polarized light through the optical microscope (50× objective). The image size is about 150 × 80 μm.

agreement with the data from [4]. However, these bands were not observed in diluted $Co_cMg_{1-c}O$ samples with $c \le 0.8$ down to 6 K, suggesting that the presence of defects, magnesium ions, destroys the long-range magnetic order (probed by AFMR) already at small dilution.

The composition dependence of Raman scattering in $Co_cMg_{1-c}O$ solid solutions at $T = 10$ K is shown in Fig. 2. Three sharp peaks due to the Brillouin zone center ($k = 0$) magnon excitations at 145, 229, and 305 cm^{-1} are well visible in pure CoO ($c = 1$), two peaks at 94 and 238 cm^{-1} remains for $c = 0.9$, and no peaks are observed upon further dilution. Note that the results for pure CoO are in agreement with the data from [5, 6]. The broad band at 400–600 cm^{-1} is attributed to first-order Raman scattering by longitudinal phonons in the Brillouin zone center [7] and is visible for $c = 1.0$ and 0.9. For $c = 0.75$, another band grows up at 300–450 cm^{-1} and the former one begins to shift to higher frequencies. Finally, two separate bands at 250–450 and 500–700 cm^{-1} are observed for $c = 0.5$. They can be attributed to transverse and longitudinal phonons in the Brillouin zone center, respectively [7]. The phonon bands vary in position due to the change in the solid-solution lattice parameter, which is larger in pure CoO ($a = 4.2615$ Å)

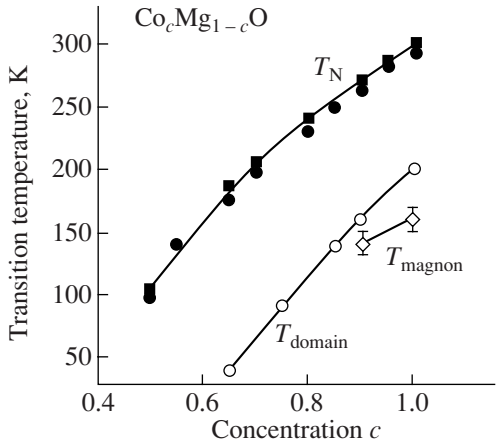

Fig. 5. Magnetic phase diagram of $Co_cMg_{1-c}O$ solid solutions: the Néel temperature (T_N) as determined from magnetic neutron diffraction [1] (solid squares) and magnetic susceptibility measurements [2] (solid circles); the temperature T_{domain} below which a domain structure exists (open circles); the temperature T_{magnon} below which magnetic excitations are observed in Raman scattering and far IR optical absorption (open diamonds).

than in MgO (a = 4.2113 Å) [1, 8]. The pronounced compositional disorder at c = 0.5 is responsible for some increase in the Raman band intensity.

The temperature dependence of Raman scattering in CoO and $Co_{0.9}Mg_{0.1}O$ solid solutions is shown in Fig. 3. The magnon-related bands decrease gradually in intensity with increasing temperature and disappear completely at about 160 K for c = 1.0 and at 140 K for c = 0.9. At the same time, the phonon-related band at 400–600 cm^{-1} remains nearly unchanged. Note that the first magnon peak, located at 92 cm^{-1} below 80 K, splits into peaks located at 75 and 91 cm^{-1} above 80 K for c = 0.9, while it only broadens for c = 1.0. The origin of the splitting is not clear.

Antiferromagnetic domain structure was observed in single-crystal $Co_{0.9}Mg_{0.1}O$ using polarized light in the temperature range up to about 160 K (Fig. 4). The twin domains have widths of the order of 0.5–2.0 μm and extend over tens of micrometers. One can see that the presence of large defects on the surface of the sample do not influence the domain structure. This suggests that the observed domain pattern reflects the bulk domain structure.

In Fig. 5, the temperature dependence of the magnetic order in $Co_cMg_{1-c}O$ solid solutions is compared for several experimental techniques. One can see that the region of magnetic ordering as determined from the domain structure, Raman scattering, and AFMR exists well below the Néel temperature (T_N), estimated from neutron diffraction [1] and magnetic susceptibility [2] measurements. This fact can be explained by different magnetic interaction correlation lengths for these experimental techniques.

4. CONCLUSIONS

We have studied the influence of dilution by magnesium ions on the magnetic order in single-crystal $Co_cMg_{1-c}O$ solid solutions. It has been observed that the twin domain structure and the contribution from magnetic excitations to Raman scattering and AFMR disappear well below the Néel temperature, determined from neutron diffraction and magnetic susceptibility measurements.

ACKNOWLEDGMENTS

This work was supported by the Latvian Government (grant nos. 05.1718 and 05-1717), the Estonian Science Foundation (grant no. 5864), and the EU TARI "EDAAM" project (contract RII3-CT-2004-506078).

REFERENCES

1. A. Z. Menshikov, Yu. A. Dorofeev, and N. A. Mironova, Solid State Commun. **98**, 839 (1996).
2. A. Arkhipov, Latv. PSR Zinat. Akad. Vestis, Fiz. Teh. Zinat. Ser. (Izv. Akad. Nauk Latv. SSR, Ser. Fiz. Tekh. Nauk), No. 2, 44 (1981).
3. W. Jauch, M. Reehuis, H. J. Bleif, F. Kubanek, and P. Pattison, Phys. Rev. B: Condens. Matter **64**, 052 102 (2001).
4. M. R. Daniel and A. P. Cracknell, Phys. Rev. **177**, 932 (1969).
5. R. R. Hayes and C. H. Perry, Solid State Commun. **14**, 173 (1974).
6. H. Chou and H. Y. Fan, Phys. Rev. B: Solid State **13**, 3924 (1076).
7. J. Sakurai, W. J. L. Buyers, R. A. Cowley, and G. Dolling, Phys. Rev. **167**, 510 (1968).
8. A. Kuzmin, N. Mironova, and J. Purans, J. Phys.: Condens. Matter **9**, 5277 (1997).