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2002 J. Phys.: Condens. Matter 14 11173

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Pressure effects on first-order magnetic Raman scattering in NiO

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Received 25 June 2002

Published 25 October 2002

Online at stacks.iop.org/JPhysCM/14/11173

Abstract

The pressure dependence of first-order magnetic Raman peak of NiO single crystal was studied up to 20 GPa at room temperature. At ambient pressure, an unknown peak is also observed at nearly the same position as the one-magnon one and their separation becomes remarkable with increasing pressure. Pressure coefficients of the one-magnon peak and the other peak are obtained as 0.4 and 1.5 cm⁻¹ GPa⁻¹, respectively. The next-nearest-neighbour antiferromagnetic exchange constant J_2 is obtained as a function of the lattice constant.

1. Introduction

NiO is known as an antiferromagnetic Mott insulator with a Néel temperature (T_N) of 523 K. The crystal structure is a cubic rock-salt one (B1) above that temperature. Below T_N , it shrinks slightly in the directions of [111] and becomes a contracted rhombohedral structure which is stable even at 141 GPa [1]. Infrared absorption, two-magnon Raman scattering [2, 3] and neutron scattering studies [4] were performed and magnon dispersion curves were obtained. In the high-pressure region, Massey *et al* [5] studied two-magnon Raman scattering up to 30 GPa and obtained the pressure dependence of the antiferromagnetic exchange constant under several assumptions. Lockwood *et al* [6] observed a first-order magnetic Raman peak around 40 cm⁻¹. They studied its temperature dependence and showed that the peak intensity at room temperature is rather stronger compared with liquid helium temperature, which is the opposite tendency to that shown by two-magnon peak. Grimsditch *et al* [7] showed unexpected polarization behaviour of this peak. However, there have been no high-pressure studies of the first-order Raman peak.

2. Experimental details

The pressure dependence of the first-order magnetic Raman peak of NiO single crystal was studied up to 20 GPa at room temperature. The samples were pressurized using a DAC

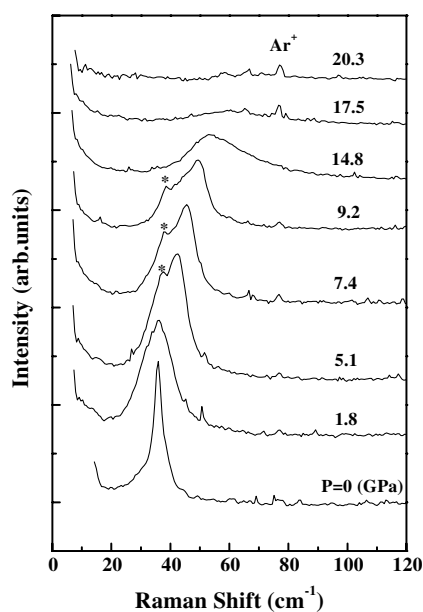


Figure 1. Raman spectra of NiO under various pressures.

(diamond anvil cell). A methanol–ethanol (4:1) solution was used as a pressure medium. The pressure calibration was performed by the standard ruby fluorescence method. Focusing of a 514.5 nm light from an argon ion laser on the sample and collection of the scattered light were performed using a Mitsutoyo FS60 microscope. The optical signal was detected by a Jobin–Yvon triple monochromator T64000 and a Jobin–Yvon Spex Spectrum-one CCD (charge coupled device) system.

3. Results and discussion

Raman spectra obtained under various pressures are illustrated in figure 1. Here a peak at 76.9 cm^{-1} is a plasma line from argon ions and we used it as a standard for wavenumber calibration. In the spectrum obtained at ambient pressure ($P = 0$), a sharp peak with a tail toward lower energy is seen around 35 cm^{-1} . We decomposed this peak into two components and obtained their peak positions as 33 and 36 cm^{-1} , respectively. These values are nearly the same as those for the one-magnon frequency at room temperature [7]. The former peak is sensitive to pressure and its Raman shift exceeds that of the latter even on slight compression. With increasing pressure, separation of these peaks becomes clear. Sudden increase in peak width is seen around 10 GPa, which may be due to the inhomogeneity of pressure resulting from the solidification of the pressure medium which occurs around this pressure. The pressure shift of the peak marked by the star is small and it vanishes around 15 GPa; however, the other one survives up to around 20 GPa. Since one-magnon Raman scattering reflects long-range magnetic order, it is possible to say that the disappearance of the one-magnon Raman peak resulted from a partial disturbance of the long-range magnetic order occurring under pressure.

Figure 2 shows the relation between peak positions and applied pressure. The pressure coefficients of the peaks are obtained as 1.5 and $0.4\text{ cm}^{-1}\text{ GPa}^{-1}$, respectively. If the magnon energy increases with the former ratio ($1.5\text{ cm}^{-1}\text{ GPa}^{-1}$), the antiferromagnetic exchange

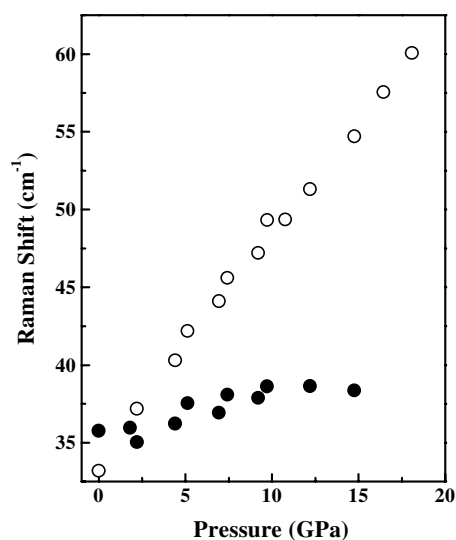


Figure 2. The pressure dependence of the magnon Raman shift. The pressure coefficients of the peaks are 1.5 (open circles) and $0.4 \text{ cm}^{-1} \text{ GPa}^{-1}$ (solid circles).

constant at 20 GPa calculated under the conditions mentioned later becomes about three times the value for ambient pressure. Since the lattice constant shrinks only by a few per cent [1], it is too large. Therefore we regard the origin of the pressure-insensitive peak (solid circles) as a one-magnon one; on the other hand, that of the other (open circles) is not clear.

From the pressure dependence of the one-magnon peak position and the pressure dependence of the lattice constant [1], the next-nearest-neighbour antiferromagnetic exchange constant J_2 is obtained as a function of lattice parameter a_0 under the following approximations. Since, in NiO, the nearest-neighbour exchange constant J_1 is much smaller than J_2 , we neglect it. As the rhombohedral axial ratio c/a is nearly the same as that of cubic B1 structure (NaCl type) even under high pressure [1], we regard the crystal structure as B1 with lattice constant a_0 . We also neglect the pressure dependence of the anisotropy energy E_A . Using $E_A = 1 \text{ cm}^{-1}$ and $J_2(P = 0) = 150 \text{ cm}^{-1}$ obtained from neutron scattering at ambient pressure [4], a relation between J_2 and a_0 is obtained as illustrated in figure 3. Here solid squares (run 1) and circles (run 2) are present data and open circles are the results from two-magnon data obtained by Massey *et al* [5]. The lattice constant dependence of J_2 obtained for the one-magnon case shows very good agreement with that for the two-magnon case.

4. Conclusions

The pressure dependence of the one-magnon Raman scattering in NiO single crystal was studied up to 20 GPa at room temperature. At ambient pressure, two peaks are observed at nearly the same position and their separation becomes remarkable with increasing pressure. Their pressure coefficients are 1.5 and $0.4 \text{ cm}^{-1} \text{ GPa}^{-1}$, and the latter is assigned to the one-magnon peak. The next-nearest-neighbour antiferromagnetic exchange constant J_2 is obtained as a function of lattice constant. The result shows a very good agreement with previous work.

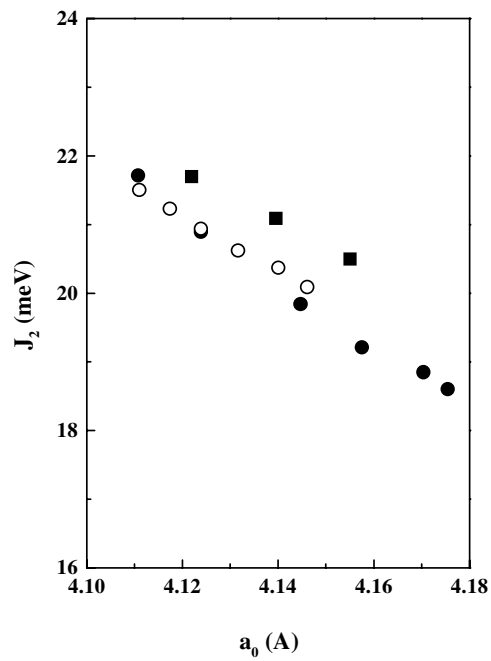


Figure 3. A relation between the lattice constant and antiferromagnetic exchange constant J_2 . The lattice constants were obtained from [1].

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