Proc. Int. Symposium on Advances in Neutron Scattering Research, Tokai, 2000
J. Phys. Soc. Jpn. **70** (2001) Suppl. A pp. 97–99

Spin Dynamics in NiO near and above the Néel Temperature

Yoshiei TODATE

Physics Department, Ochanomizu University, 2-1-1 Bunkyo-ku, Otsuka, Tokyo 112-8610, Japan

Measurements of the inelastic magnetic scattering from a single crystal NiO have been carried out in order to obtain the paramagnetic $\chi(q)$ i.e., the spin correlation above the Néel temperature. It has been found that rather intense paramagnetic scattering exists in the energy region of $\hbar\omega \sim 20$meV and is almost unchanged up to T=623 K. The intensity is concentrated near the antiferromagnetic zone center, indicating the existence of the significant instantaneous antiferromagnetic correlations above $T_{\rm N}$. Another strongly temperature-dependent inelastic scattering, which shows the critical behavior, has been found at $\hbar\omega \lesssim 4$meV.

KEYWORDS: NiO, spin correlation, paramagnetic scattering, critical scattering

§1. Introduction

Since NiO is often considered to be a prototype of the insulating $3d$ transition metal oxides with large electron correlations, there have been extensive investigations of its electronic structure.[1] For analyses of magnetic properties of NiO, on the other hand, a Heisenberg spin-only hamiltonian has conventionally been assumed because the insulating gap and the local magnetic moments persist in the paramagnetic state. Magnetic excitations, for example, at the ordered state were analyzed by using the linear spin wave theory.[2] It has recently been shown, however, that the similar inconsistency to NiO of the exchange parameters exists also in the antiferromagnetic ordered complex perovskites[3] (e.g., Ba_2NiWO_6 which show the same type-II antiferromagnetic (AF) ordering on the FCC lattice); the exchange parameters obtained from the high temperature susceptibilities significantly differ from those determined from the spin-waves. Because the discrepancy is too large to attribute entirely to the shortcomings of the molecular field approximation and, more interestingly, because of the remarkable similarity between NiO and Ba_2NiWO_6, some particular situation of the FCC lattice has been suggested. In connection to the electron correlation effect, further investigations will be interesting to see whether the "simplest" localized spin model is sufficient to describe microscopic behavior of spins in these compounds for all the temperature range including critical and paramagnetic regions. The motivation of this study is that, in our view, some visible effects of the magnetic phase transition (accompanied by the symmetry braking) on the electron correlation could be expected in the microscopic spin dynamics in these coumpounds.

The above-mentioned discrepancy is expressed in terms of the wavevector-dependent susceptibility[4] $\chi(\boldsymbol{Q}) = g^2\mu_B^2 S(S+1)/\{3kT - 2S(S+1)J(\boldsymbol{Q})\}$. For the uniform susceptibility $J(\boldsymbol{Q}=0) = 12J_1 + 6J_2$, whereas for the staggered susceptibility which diverges at the Néel temperature ($T_{\rm N}$), $J(\boldsymbol{\tau}_0) = -6J_2$, where J_1 and J_2 are 90° and 180° interactions, respectively. Although no significant contribution of J_1 to the spin wave excitations is observed, the uniform susceptibilities always give larger $J(0)$ (or stronger AF correlation) than expected from J_2 only. It seems essentially important to reveal the origin of this inconsistency because it is related to the interaction process of $3d$-electrons in NiO or to unknown effects yet to be considered. The chief aim of this study is to verify the existence or nonexistence of the additional correlation between nearest neighbor spins coupled via J_1 in the paramagnetic state. We note that the finite spin correlation via J_1 in the paramagnetic state may, if exists, be caused by the symmetry change effect on the electron correlation and that the electronic and magnetic properties of NiO depend essentially on J_1 because of its frustrating nature in the FCC lattice.

For this purpose, a straightforward way to see the paramagnetic spin correlation is to obtain $S(Q)$ or measure the q-dependence of the susceptibility; we point out that at $T > T_{\rm N}$ and for small q, $\chi(\boldsymbol{\tau}_0+\boldsymbol{q})$ can be written as

$$\chi(\boldsymbol{\tau}_0+\boldsymbol{q}) \simeq \frac{g^2\mu_B^2 S(S+1)}{3kT_{\rm N}} \times \frac{1}{r_1^2\{\kappa_1^2 + (q^2 + 2\frac{J_1}{J_2}(q_xq_y + q_yq_z + q_zq_x))\}} \quad (1.1)$$

for one of four T-domain patterns in NiO, where a is the lattice parameter of the chemical cubic unit, $r_1^2 = a^2/6$ and $\kappa_1^2 = r_1^{-2}(T - T_{\rm N})/T_{\rm N}$. In the paramagnetic state except in the vicinity of $T_{\rm N}$, however, it may be more natural to express in an averaged form as,[5]

$$\chi(\boldsymbol{\tau}_0+\boldsymbol{q}) \simeq \frac{g^2\mu_B^2 S(S+1)}{3kT_{\rm N}} \frac{1}{r_1^2(\kappa_1^2 + (1+\frac{J_1}{J_2})q^2)} \quad (1.2)$$

As the above expression for the q-dependent susceptibility explicitly involves the contribution of J_1, the paramagnetic scattering experiment is expected to provide substantial information. For the purpose of deducing this $\chi(\boldsymbol{\tau}_0+\boldsymbol{q})$ by integrating $S(\boldsymbol{Q},\omega)$ and obtaining the information on the paramagnetic spin correlations, we have measured the inelastic magnetic scattering from a single crystal of NiO. It has also been required to determine the energy scale of the dynamics because the experimental difficulty may become crucial due to the high energy transfer.

§2. Experimental

The measurements were carried out by using pulsed neutrons with the MAX spectrometer[6] at KENS in a temperature range from room temperature to 623 K (1.17 T_N). A single crystal of about 20g in weight was mounted in a infrared furnace. The transition temperature of this crystal was determined to be 533 K from temperature variation of the AF Bragg intensity. The time-of-flight (TOF) scans were made along the [110] direction through the $(\frac{1}{2},\frac{1}{2},\frac{3}{2})$ point and along [11-1] through $(\frac{3}{2},\frac{3}{2},\frac{1}{2})$.[7]

Examples of the inelastic spectra obtained in different energy regions are shown in Figs.1, 2 and 3. Figure 1 shows the temperature dependence of the excitation observed at about $\hbar\omega$=20meV. Note that the scattering intensities shown in the figure are not either constant-q nor constatnt-E spectra because these TOF scans are made with fixed scattered neutron energies and fixed scattering angles, and therefore the energy transfer varies as a function of Q, although the scans are made along the high-symmetry directions. (For more information on this experimental technique, see Ref. 6.) The dotted line shows the calculated spin-wave peak profile using the exchange parameters determined by Hutchings and Samuelsen at 78 K.[2] The data clearly show the intense inelastic scattering concentrated around the AF zone center above T_N, indicating the significant instantaneous AF correlation. Note that the paramagnetic scattering peak is roughly unchanged up to T =623 K. Below 503 K, an influence of the temperature renormalization on the spin-wave energy is not significant in this energy region. It is shown in Fig. 2, on the other hand, that the excitation in the higher energy region ($\sim$ 45meV) is rather damped in the paramagnetic state. At the present time, unfortunately, the data are not sufficient to cover wide enough $S(\boldsymbol{Q},\omega)$-region for deriving complete $\chi(\boldsymbol{\tau}_0 + \boldsymbol{q})$.

Another newly found feature is the strongly temperature-dependent inelastic scattering around $\hbar\omega \sim$ 4meV. This scattering shows a maximum at T_N as shown in Fig. 3. A similar low energy inelastic peak observed at a different AF zone (but the zone which belongs to the same domain structure) can also be seen in Fig. 2. The intensity of this component exhibits typical critical behavior as plotted in the inset of Fig. 3. A single crystal experiment carried out early on by Roth showed no clear magnetic diffuse scattering neat T_N.[8] However, the present study has clearly shown that the critical fluctuation which contributes to the total scattering is mostly inelastic.

§3. Discussion

The present measurements have shown that the distinct paramagnetic scattering exists in the energy region of $\hbar\omega \sim$ 20meV and is almost unchanged up to T=623 K. Since the intensity is concentrated around the AF zone center, it clearly indicates the existence of the significant instantaneous AF correlations above T_N. This paramagnetic response would be an important characteristic of NiO. Higher energy excitations ($\hbar\omega \gtrsim$ 40meV) broaden more rapidly above T_N. It is also suggested from these results that the energy integration up to at least $\sim$ 30meV is necessary in order to obtain a reliable instantaneous spin correlation function. At present, the data are not sufficient to verify whether there is an additional contribution from the nearest neighbor AF correlation to this dynamical correlation. It will be the future study to obtain a complete map of $S(\boldsymbol{Q},\omega)$. It is also interesting to compare again with results on Ba_2NiWO_6.

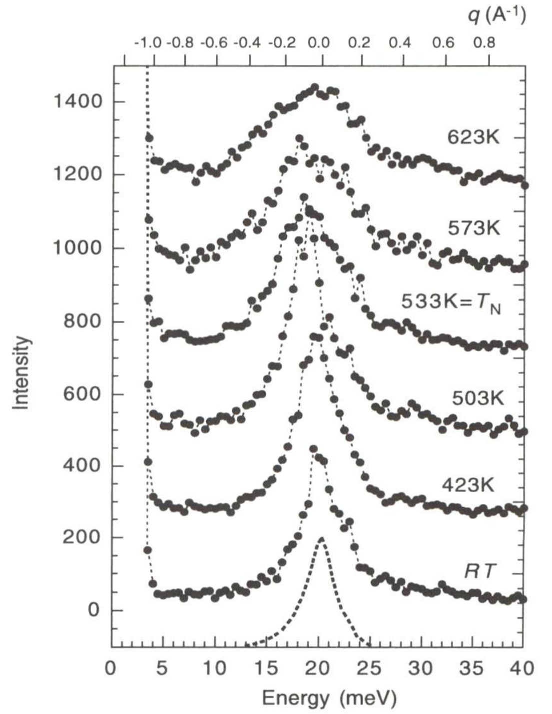

Fig. 1. Temperature variation of the excitation spectrum around (3/2,3/2,1/2). Note that the energy transfer varies as a function of Q because of the TOF method with fixed final energy. However, These data roughly represent the constant-E spectra because the peaks are relatively sharp. The dotted line drawn below shows the spin-wave peak convoluted with the instrumental resolution.

As regards the low energy inelastic components which show the critical behavior near T_N, together with the reported data showing no conventional Lorentzian-shaped diffuse scattering, one of possible explanations would be some sort of sublattice (domain) switching. Because of the particular situation of type-II AF ordering in the FCC lattice where the incompatible four types of T-domains are equally populated, it is natural to consider that all the finite T-domains are equivalent and switching temporally and spatially near T_N. Furthermore, the fluctuation should be coupled with the lattice distortion. The data suggest rather rapid fluctuation with a time scale of $\hbar\tau^{-1} \sim$ 3meV. It should be noted that the fluctuation or the dynamical disorder remains at room temperature as shown in Fig. 3. If this is the case, a study of this lower energy fluctuation belongs to a somewhat different subject from the electron correlation effect in

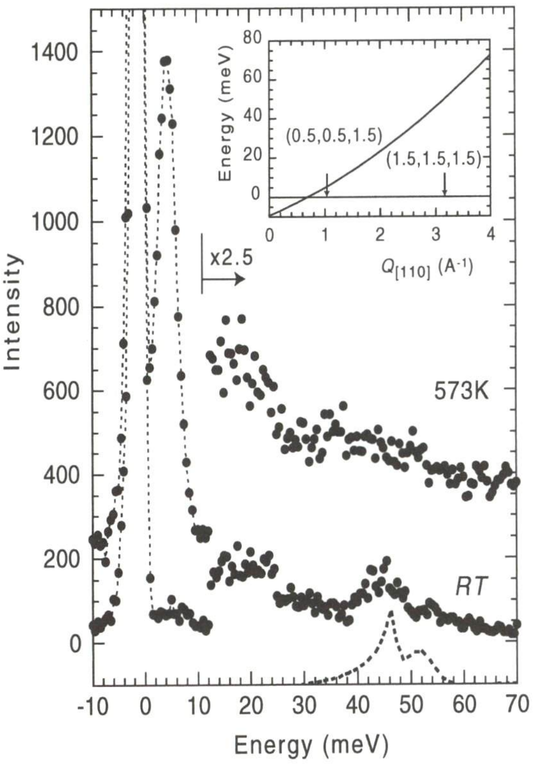

Fig. 2. Example of a spectrum scanned along [110]. The spin wave excitation observed at about 45meV belongs to the zone $(\frac{3}{2}, \frac{3}{2}, \frac{3}{2})$ while the low energy portion scans the zone $(\frac{1}{2}, \frac{1}{2}, \frac{3}{2})$ as shown by a TOF locus in the inset. Note the low energy intense peak at T=573 K.

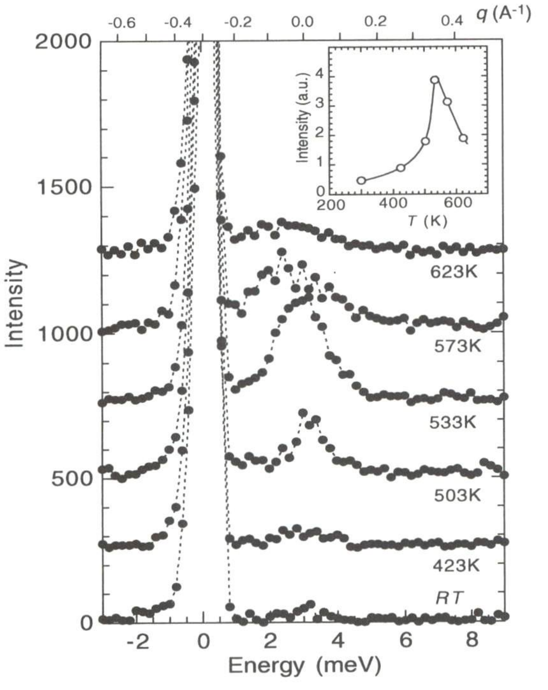

Fig. 3. Temperature variation of the low energy inelastic scattering measured around $(\frac{3}{2}, \frac{3}{2}, \frac{1}{2})$. The integrated intensity in a range of 1.6-6meV is shown in the inset.

NiO, but it will also be interesting and important.

Acknowledgements

The author would like to thank Prof. K. Kohn for providing a single crystal sample and also for his useful discussions. The author also wishes to thank Prof. S. Ikeda for help of obtaining another single crystal.

1) S. Hufner : Adv. Phys. **43** (1994) 183; B. Brandow : Adv. Phys. **26** (1977) 651.
2) M. T. Hutchings and E. J. Samuelsen : Solid State Commun. **9** (1971) 1011.
3) Y. Todate : J. Phys. Chem. Solids **60** (1999) 1173.
4) W. Marshall and R. D. Lowde : Rep. Progr. Phys. **31** (1968) 705.
5) The situation where half of 12 nearest neighbors are coupled antiferromagnetically in the average is considered.
6) Y. Todate, K. Tajima, H. Ikeda and S. Tomiyoshi : Jpn. J. Appl. Phys. **29** (1990) 1220.
7) The scan was slightly shifted in order to avoid strong (111) nuclear Bragg scattering.
8) W. L. Roth : Phys. Rev. **111** (1958) 772.