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CRITICAL BEHAVIOUR OF NiO AS AN ANTIFERROMAGNET WITH NONRIGID LATTICE*)

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Abstract: We discuss the experimental results obtained for NiO near the Neel temperature by the neutron diffraction method. Since NiO has a noticeable exchange induced distortion we compare the experimental results with the molecular field theory and the Green function approximation proposed for magnets with such effects. However, we found that the agreement with these two theories is by far worse than the agreement with the low-temperature and high-temperature series expansions predictions for the Ising and Heisenberg models on rigid lattices. This finding should stimulate study of the Ising and possibly the Heisenberg models on nonrigid lattices.

1. Introduction

Nickel-oxide is almost ideal antiferromagnet of the second kind[1]. Spins of the ions Ni^{2+} in the plane (111) of the magnetic lattice are ordered ferromagnetically below the Neel temperature $T = 523$ K. The dominant interaction between these spins is the nearest-neighbour exchange interaction[2]. The intensity of the

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magnetically diffracted neutrons on the planes (111) of NiO depends on the temperature of the sample according to the relation

$$I_m(T) = \text{const.}\, F_m^2(T) < \hat{\vec{S}}_i \cdot \hat{\vec{S}}_j > T, \tag{1}$$

where F_m is the magnetic structural factor of the ion Ni^{2+}, and $< \hat{\vec{S}}_i \cdot \hat{\vec{S}}_j >_T$ is the correlation function of the spins situated on the i-th and j-th site of the magnetic lattice. F_m depends on temperature mainly through the Debye-Waller factor, as the form-factor is practically temperature independent[3]. Since NiO has high melting temperature and relatively low Debye temperature, the Debye-Waller factor should exhibit a small temperature change in the interval between the room temperature and T_N. Our measurements show that the intensity $I_m(T)$ decreases less than 2 percent when temperature increases from 91 K up to 572 K. Therefore, we conclude according to the relation (1) that $I_m(T)$ predominantly depends on the spin correlations.

In the molecular-field approximation (MF) the following relation may be accepted

$$< \hat{\vec{S}}_i \cdot \hat{\vec{S}}_j >_T \cong < S_i >_T^2 \equiv \bar{S}(T)^2, \tag{2}$$

where $\bar{S}$ is the average spin at the temperature T. Lines[2] argued that this relation is plausible in the case of the nearest-neighbour interaction.

The order parameter we are concerned with is the normalized average spin in the (111) plane of the NiO lattice

$$M(T) = \frac{< \hat{\vec{S}} >_T}{< \hat{\vec{S}} >_0}. \tag{3}$$

Accepting $F_m^2 = \text{const.}$ in Equ. (1) and using Equ. (2) it follows

$$M(T) = \sqrt{\frac{I_m(T)}{I_m(0)}}, \tag{4}$$

where $I_m(0)$ is usually obtained by extrapolating the function $I_m(T)$ at 0 K. Therefore, by measuring the intensity $I_m(T)$ we have obtained the order parameter $M(T)$ as a function of temperature. Firstly, we will discuss the experimental method and secondly, we will compare our results with the following theoretical predictions:

- molecular field approximation,
- the Green function calculations, and
- the series expansions results for the Ising and Heisenberg models.

2. Experiment

The measurement of the function $I_m(T)$ has been done with the neutron spectrometer presented in Fig 1. Pyrolitic graphite used as the neutron monochromator gives the following set of neutron wavelenghts 4.16, 2.08, 1.39, 1.04 and 0.38 Å, while the time-of-flight equipment provides the simultaneous observation of the Bragg reflections of each wavelenght. NiO has the fcc lattice with the lattice constant $a = 4.1765$ Å[4], whereas its magnetic lattice has a twice as lare elementary cell[1]. The scattering angle was chosen in order to satisfy the Bragg law for the reflection of neutrons ($\lambda = 4.16$ Å) on the (111) planes of the magnetic lattice so that the higher order reflections occurred at the monochromator would satisfy the Bragg laws for the same order reflections on the sample[5].

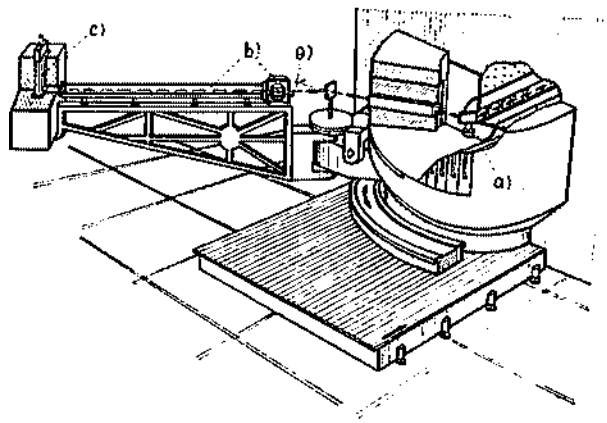

Fig. 1. Neutron spectrometer

a) Monochromator – pyrolitic graphite,
b) Chopper and time-of flight tube,
c) He^3 – neutron detector,
Θ Scattering angle.

Fig. 2 represents the spectrum of the diffracted neutrons on the (111) planes for the temperatures below (458 K) and above (531 K) the Neel point (523 K). One can notice the disappearance of the magnetic Bragg peaks (111) (except for the contribution of the incoherent elastic scattering of neutrons) and no change with the temperature of the (222) and (444) nonmagnetic peaks. We have chosen the results obtained for the peak that corresponds to the (111) plane and neutrons of 4.16 Å to compare with the theoretical predictions. The measured intensity $I_m(T)$ has been corrected with the factor

$$\frac{I_{\infty}}{I_m(T)},$$

where $I_n(T)$ is the intensity of the nonmagnetic peak (444) and $\bar{I}_n$ is the corresponding mean value which has been independently estimated for the temperature ranges between 91 K and 447 K and between 457 K and 572 K in order to take into account the Debye-Waller factor. This correction has to eliminate the fluctuation effects caused by uncontrolable change of experimental conditions in the reactor hall.

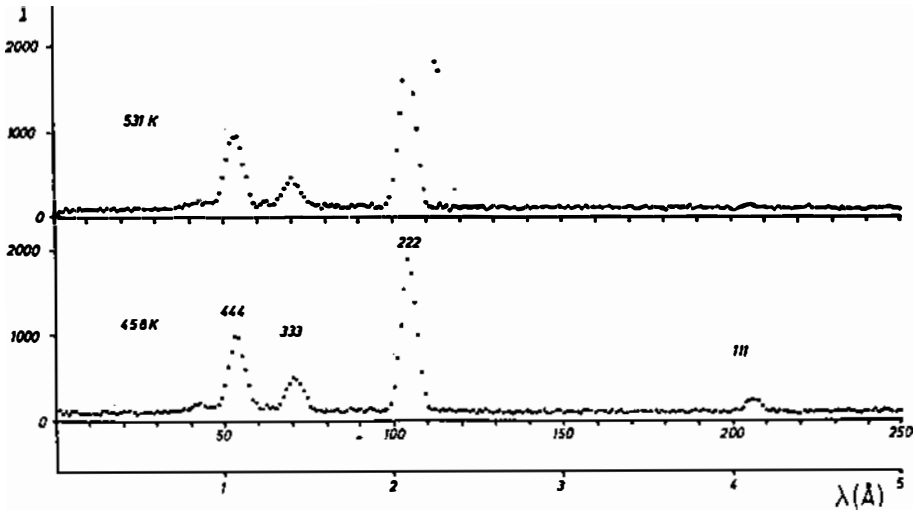

Fig. 2. Comparison of the diffraction patterns at $T = 458$ K and $T = 531$ K.

3. *Comparison with theory*

The experimental results for the spontaneous magnetization obtained according to Equ. (4) are compared with the mean field predictions[6], with the Green function calculations[6] and with the numerical results for the Ising and Heisenberg models.

The mean field theory for a distortionable lattice, originally derived for MnO, predicts the following behaviour for the spontaneous magnetization

$$\bar{S}(T) = SB(x), \tag{5}$$

where $B(x)$ is the Brillouin function, with

$$x(T) = \frac{G\mathcal{J}_2 S}{kT}\bar{S}(T)\left[1 + \frac{j}{\mathcal{J}_2}\bar{S}^2(T)\right]. \tag{6}$$

The Green function approximation[6] derived also for the lattice with noticeable distortion effects provides a more complicated formula for the spontaneous magnetization

$$x(T) = S \ln \frac{Y(T)+1}{Y(T)-1},$$
$$Y(T) = \frac{2}{N} \sum_{\vec{K}} A(\vec{K}) \operatorname{cth} \frac{E_0(\vec{K}) \cdot \bar{S}(T)}{2kT}. \tag{7}$$

The funcions E_0 and A are defined by

$$E_0 = \sqrt{u^2 - v^2}, \qquad A = \frac{u}{E_0},$$

where

$$u + v = 4\mathcal{J} \sum_{\alpha \neq \beta}^{3} c_\alpha c_\beta + 4\mathcal{J}_2 \sum_{\alpha}^{3} c_\alpha + 4j\, S\,[3 - \sum_{\alpha \neq \beta}^{3} s_\alpha s_\beta],$$
$$u - v = 4\mathcal{J}_1 \sum_{\alpha \neq \beta} s_\alpha s_\beta + 4\mathcal{J}_2 \sum_{\alpha} s^2 + 4j\, S\,[3 - \sum_{\alpha \neq \beta} c_\alpha c_\beta], \tag{8}$$
$$s_\alpha = \sin(K_\alpha a), \qquad c_\alpha = \cos(K_\alpha a), \qquad \alpha, \beta = x, y, z.$$

$\mathcal{J}_1$ and $\mathcal{J}_2$ are the nearest and next-nearest neighbour exchange parameters respectively, whereas j is the parameter which is responsible for the deformation effects caused by the exchange mechanism[7].

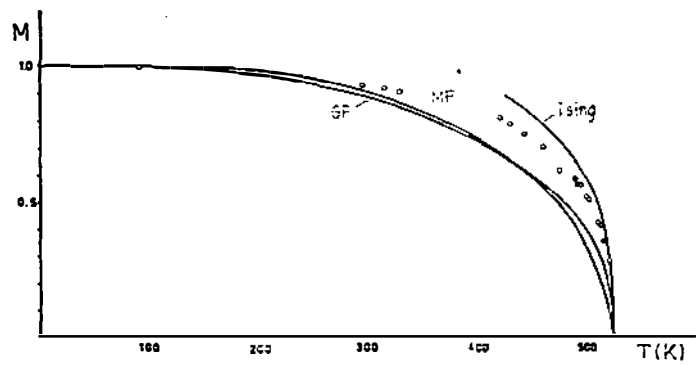

Fig. 3. The comparison of the experimental results with the Ising model, the mean field theory (MF) and Green function calculations (GF).

Comparison of the experimental results with the theoretical expectations (5) and (7) is presented in Fig. 3, where we have used the following values $S = 1$,

$\mathcal{J}_2 = 1.8 \cdot 10^{-14}$ erg and $\frac{j}{\mathcal{J}_2} = 0$ in the case of molecular field formula (5), and $S = 1$, $\mathcal{J}_1 = 1.973 \cdot 10^{-14}$ erg, $\mathcal{J}_2 = 2.953 \cdot 10^{-14}$ erg and $\frac{j}{\mathcal{J}_2} = 0.0014$ in the Green function formula (7). The quoted values follow the experimental results for the critical temperature[5,8] and elastic constants[8], and the corresponding theoretical formulas[5,7]. It may seem surprising that in (5) we have used $\frac{j}{\mathcal{J}_2} = 0$ instead of $\frac{j}{\mathcal{J}_2} = 0.0016$ which would be the value consistent with the molecular field theory and experimental findings. However, such a small value of the ratios $\frac{j}{\mathcal{J}_2}$ has almost no influence on the behaviour of the related curve in Fig. 3, whereas in the Green function case this influence is noticeable.

We can observe in Fig. 3 a disagreement between the experimental results and calculations based on the mean field and the Green function theory[5,6]. One could produce a kind of agreement between the two groups of results by using some larger values of the ratio $\frac{j}{\mathcal{J}_2}$, in formulas (5) and (7). But these values would be inconsistent with the corresponding experimental findings[8] and thus the possibly achieved agreement would be void in physical content.

In Figs. 3 and 4 we present the comparison of the experimental results and numerical predictions for the Ising and Heisenberg models with the nearest-neighbour interactions and with rigid lattices. The comparison emphasizes estimations of the critical point exponent and the appropriate critical amplitude B[10], i. e. here we assume that near the critical point spontaneous magnetization vanishes as

$$M = B\left(\frac{T - T_c}{T_c}\right)^{\beta}. \tag{9}$$

In the case of the Ising model we have used estimations $B = 1.488$ and $\beta = 0.3125$ obtained by Essam and Hunter[11]. In the case of the $S = \infty$ Heisenberg model, Stephenson and Wood's estimation[12] $B = 1.405$ and $\beta = 0.38$ have been used. For $S = \frac{1}{2}$ Heisenberg model we considered both possibilities $\beta = 0.35$ and $\beta = 0.385$ (Refs.[13,14]) and corresponding estimation $B = 1.481$ and $B = 1.688$ obtained from Milošević and Stanley[15].

It can be noticed from Figs. 3 and 4 that both the Ising and Heisenberg model calculations provide a better agreement with the experimental results than the mean-field theory and the Green function approximation. We should stress that the latter two were proposed for a magnet which has lattice with noticeable distortion, whereas the numerical predictions for the Ising and Heisenberg model were

made for rigid lattices. Therefore, one may judiciously expect that taking into account the exchange induced distortion effects would improve the obtained agreement, which has already been demonstrated in the case of CoO which exhibits stronger distortion effects than NiO[16].

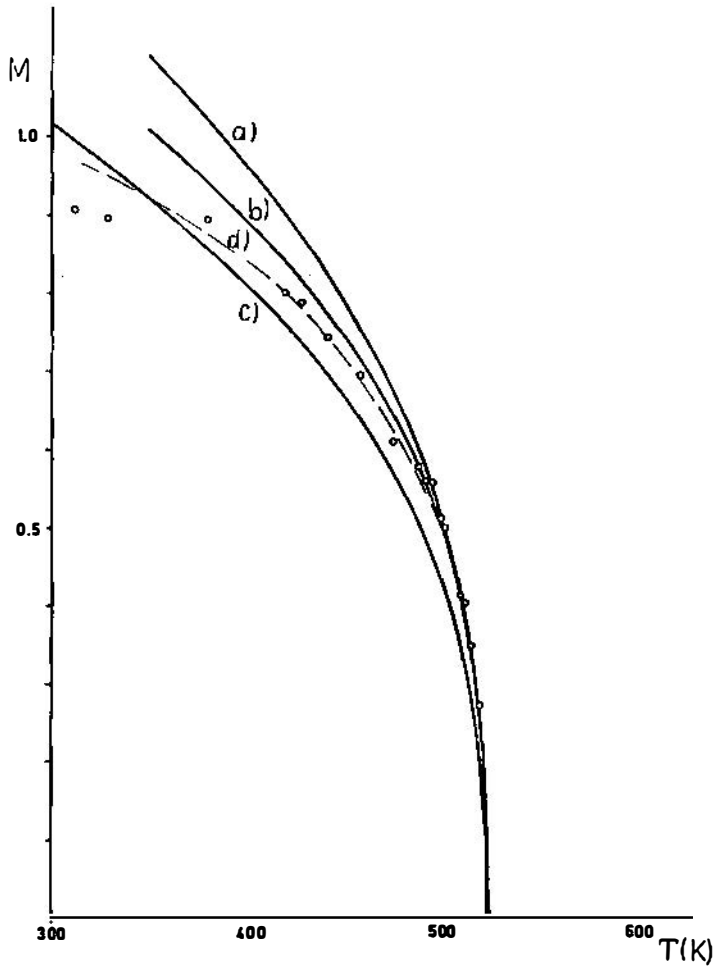

Fig. 4. The comparison of the experimental results with the numerical prediction for the Heisenberg model using the estimations:

a) $S = \frac{1}{2}$; $\beta = 0.385$; $B = 1.688$

b) $S = \frac{1}{2}$; $\beta = 0.35$; $B = 1.481$

c) $S = \infty$; $\beta = 0.38$; $B = 1.405$

d) The fitting for $\beta = 0.33$ and $B = 1.368$.

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KRITIČNO PONAŠANJE NIKL-OKSIDA KAO ANTIFEROMAGNETIKA SA NEUKRUĆENOM REŠETKOM

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Sadržaj

U ovom članku razmatrano je kritično ponašanje NiO pomoću metoda neutronske difrakcije. Pošto NiO ima izražene efekte distorzije indukovane interakcijom izmene mi poredimo eksperimentalne rezultate sa teorijom molekularnog polja i aproksimacijama koje su dobijene metodom Greenovih funkcija u slučaju magneta sa odgovatrajućim efektima.

Međutim, slaganje sa ovim dvema teorijama je gore nego podudarnost sa rezulatima dobijenim metodama niskotemperaturskih i visokotemperaturskih razvoja za Isingov i Heisenbergov model na krutim rešetkama. Ova činjenica stimuliše studiju Isingovog, a ako je moguće, i Heisenbergovog modela na neukrućenim rešetkama.