

# Magnetic susceptibilities, their temperature variation, and exchange constants of NiO

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The temperature variation of the magnetic susceptibilities of NiO for a powder sample and a single crystal has been measured from 4.2 to 700 K. The single crystal was stressed along a  $\langle 111 \rangle$  direction on cooling through  $T_N$  ( $= 524$  K) with successive stress magnitudes of 7, 30, and 42 bars, followed in each case by measurements of  $\chi_{\parallel}^*$  [ $\vec{H}$  parallel to a (111) plane with  $H = 200$  Oe] from 4.2 to 700 K. From the behavior of  $\chi_{\parallel}^*$  with stress, a stress of 30 bars was found to be sufficient to completely remove the  $T$  domains. With the use of the data of initial powder susceptibility  $\chi_p = (\chi_{\parallel} + 2\chi_{\perp})/3$  and  $\chi_{\parallel}^* = (\chi_{\parallel} + \chi_{\perp})/2$  at various temperatures, the temperature variation of  $\chi_{\parallel}$  and  $\chi_{\perp}$ , the principal susceptibilities, is determined. After correcting for  $\chi_{\text{Vv}}$  and  $\chi_d$  (the Van Vleck and diamagnetic susceptibilities) with  $\chi_{\text{Vv}} + \chi_d = 3.2 \times 10^{-6}$  cm<sup>3</sup>/g,  $\chi(T_N) = 8.8 \times 10^{-6}$  cm<sup>3</sup>/g,  $\chi_{\perp}(0) = 6.8 \times 10^{-6}$  cm<sup>3</sup>/g, and  $\chi_{\parallel}(0) = 0$  are determined. It is shown that the spin-wave theory correctly gives the ratio  $\chi(T_N)/\chi_{\perp}(0)$  and the temperature dependence of  $\chi_{\parallel}$  to about 200 K if the experimental value of the spin reduction  $\Delta S = 0.19$  is used for Ni<sup>2+</sup>. From the experimental values of  $\chi(T_N)$  and  $T_N$  and the use of random-phase-approximation Green's-function theory, the magnitudes of exchange constants for NiO, viz.  $J_1 = 34$  K and  $J_2 = 202$  K, are determined.

## I. INTRODUCTION

The nature of magnetic ordering in NiO (as well as in MnO, FeO, and CoO) became known from the early neutron diffraction studies by Roth<sup>1</sup> and from the magnetic susceptibilities by La Blanchetais<sup>2</sup> and Singer.<sup>3</sup> According to these studies, NiO is a type-2 antiferromagnet with  $T_N \simeq 523$  K, such that the moments lying in the (111) planes are parallel with the neighboring (111) planes stacked antiferromagnetically. The spin-easy axes are the three equivalent  $\langle 11\bar{2} \rangle$  directions. In these oxides, exchange-striction accompanies magnetic ordering, this being a contraction in NiO along the four equivalent  $\langle 111 \rangle$  axes in the NaCl structure of these oxides. This multiplicity in the distortion and the spin-easy axes results in the formation of  $T$  and  $S$  domains, respectively.<sup>1</sup> Consequently the measurements of the principal susceptibilities,  $\chi_{\parallel}$  and  $\chi_{\perp}$ , in these oxides is not a straightforward matter. In recent papers from this laboratory<sup>4,5</sup> we have reported the measurements of  $\chi_{\parallel}$  and  $\chi_{\perp}$  in MnO and CoO systems. This was accomplished by applying uniaxial stress along selective directions (along [111] in MnO and along [001] in CoO) as the samples are cooled through  $T_N$  so as to obtain single- $T$ -domain crystals. In this paper we report the determinations of  $\chi_{\parallel}$  and  $\chi_{\perp}$  in NiO. Earlier measurements of La Blanchetais<sup>2</sup> were limited to powder samples for temperatures above 90 K, and in the data by Singer<sup>3</sup> on single crystals,  $\chi_{\parallel}$  and  $\chi_{\perp}$  could not be determined because of the presence of  $T$  domains. In the recent torque measurements of Kurosawa *et al.*,<sup>6</sup> only the difference susceptibility  $\chi_{\perp} - \chi_{\parallel}$  could be determined. Thus the measurements reported here represent the first determination of  $\chi_{\parallel}$  and  $\chi_{\perp}$  in NiO. Temperature variation of  $\chi_{\parallel}$  and  $\chi_{\perp}$  from 4.2 to about 700 K are presented and exchange constants in NiO are reexamined in light of these new measurements.

## II. EXPERIMENTAL DETAILS

The single crystal of NiO used in these experiments was cut from a larger boule purchased from Cristal-Tec, Grenoble, France. The sample, measuring  $2 \times 1.5 \times 1.5$  mm<sup>3</sup>, was x-ray aligned. The magnetic susceptibilities were measured with the Faraday technique using a Cahn microbalance and Lewis-gradient coils,<sup>7</sup> with the absolute and relative precisions of about 1% and 0.3%, respectively. Measurements of  $\chi$  were made with a uniaxial stress applied along a [111] axis, along with the measurements on the powdered samples. The stress cell was made from oxygen-free, high-purity copper block and the stress was applied by turning a screw. A strain-gauge arrangement was used to determine the stress at room temperature. The background correction due to the stress cell at various temperatures was determined in a separate experiment. The NiO powder sample was made from the NiO single crystal and it was wrapped in high-purity Al foil for  $\chi$  measurements from 4.2 to 700 K. The  $\chi$  values were later corrected for Al background in a manner described elsewhere.<sup>7</sup>

## III. MAGNETIC SUSCEPTIBILITIES

The temperature variation of initial  $\chi_p$  ( $\chi$  for powdered sample) measured at 200 Oe is shown in Fig. 1 for the temperature range of 4.2 to 700 K. The observed  $\chi_p$  versus  $T$  behavior is similar to that of an antiferromagnet, with  $\chi_p(0) \simeq 7.75 \times 10^{-6}$  cm<sup>3</sup>/g and  $\chi_p(T_N) \simeq 12.0 \times 10^{-6}$  cm<sup>3</sup>/g. From the maximum of  $\partial(\chi_p T)/\partial T$  against  $T$ ,  $T_N \simeq 524.5$  K is determined. Our values of  $\chi_p$  are in agreement with those of Ref. 2, the latter being limited, however, to  $T > 90$  K. No noticeable field dependence of  $\chi_p$  was observed for fields up to 8 kOe.

For measurements on the single crystals of NiO, a pro-

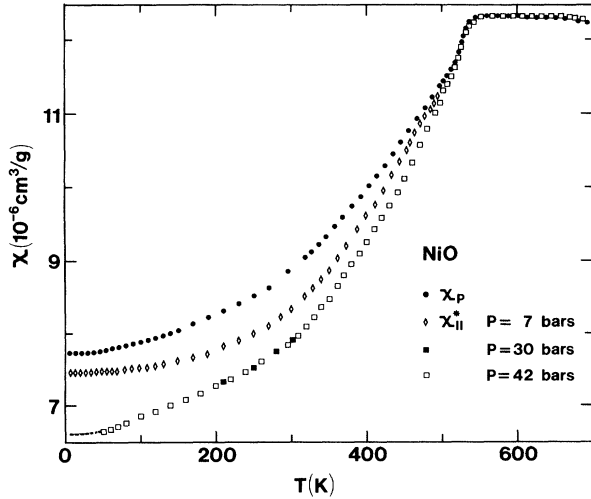


FIG. 1. Temperature dependence of the power susceptibility  $\chi_p$  measured at 200 Oe and that of  $\chi_{||}^*$  ( $\vec{H} \perp [111]$ , also at  $\vec{H} = 200$  Oe) with stress of 7, 30, and 42 bars along the  $[111]$  axis. For clarity, only a limited amount of data near  $T_N$  are shown.

cedure similar to that used in our studies on MnO (Ref. 4) was followed as the crystal and magnetic structures of MnO and NiO are similar. Uniaxial stress  $\vec{P}$  along the  $[111]$  axis was applied at room temperature, the temperature of the sample was raised to well above  $T_N$  (to about 700 K), and the sample was then cooled through  $T_N$  under the uniaxial stress. This procedure forces the crystal to contract only along the stressed  $[111]$  axis, thus eliminating  $T$  domains if sufficient stress is available. The measurements of  $\chi$  were then made from 4.2 to 700 K with increasing temperatures, in  $\vec{H} = 200$  Oe and with  $\vec{H} \perp [111]$  [or  $\vec{H} \parallel (111)$  plane]. Following our nomenclature in MnO, this  $\chi$  for  $\vec{H} \parallel (111)$  plane is termed as  $\chi_{||}^*$  and it equals  $(\chi_{||} + \chi_{\perp})/2$  because of the lack of the uniqueness of the easy axis in the  $(111)$  plane.<sup>8</sup> Thus  $\chi_{||}$  cannot be measured directly. The data for  $\chi_{||}^*$  versus temperature are also shown in Fig. 1 for  $\vec{P} = 7, 30,$  and 42 bars. It is evident that values of  $\chi_{||}^*$  for 7 bars are less than  $\chi_p$  values. Increasing the stress to 42 bars further lowers  $\chi_{||}^*$  (because of elimination of  $T$  domains). However,  $\chi_{||}^*$  values for  $\vec{P} = 30$  and 42 bars coincide, showing that under these stress values, a single- $T$ -domain crystal has been obtained. In MnO,  $\vec{P} = 25$  bars was found to be adequate to remove  $T$  domains.<sup>4</sup> Near and above  $T_N$ , no noticeable difference could be detected between the magnitudes of  $\chi_{||}^*$  and  $\chi_p$ .

The measured susceptibilities  $\chi_{||}^* = (\chi_{||} + \chi_{\perp})/2$  and  $\chi_p = (\chi_{||} + 2\chi_{\perp})/3$  can be used to derive the magnitudes of  $\chi_{||}$  and  $\chi_{\perp}$  at various temperatures.<sup>9</sup> The values so obtained at different temperatures are shown in Fig. 2 with the right-hand scale applicable. It is noted that  $\chi_{||}$  so evaluated does not approach 0 as  $T \rightarrow 0$  K. Since  $\text{Ni}^{2+}$  orbital moment is usually quenched,  $\chi_{||}(0) \approx 0$  is expected. This apparent discrepancy is due to a relatively large contribution from  $\chi_{VV}$ , the Van Vleck susceptibility which is given by

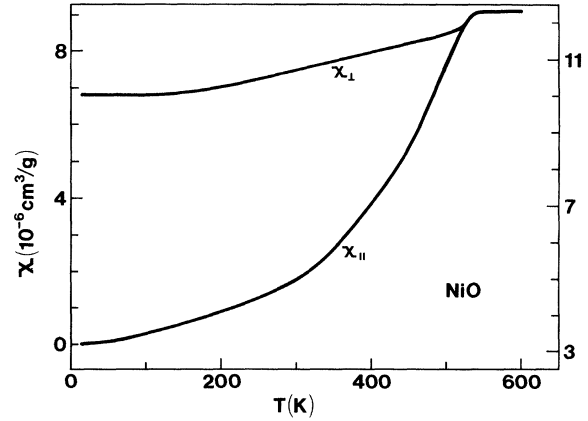


FIG. 2. Temperature dependence of  $\chi_{||}$  and  $\chi_{\perp}$ , derived from  $\chi_{||}^*$  at 42 bars and  $\chi_p$ . Right scale:  $\chi$ 's are uncorrected for  $\chi_{VV}$  and  $\chi_d$ . Left scale:  $\chi$ 's are corrected for  $\chi_{VV} + \chi_d = 3.2 \times 10^{-6}$   $\text{cm}^3/\text{g}$ .

$$\chi_{VV} = 2N\mu_B^2 \sum_{\mathcal{K}} [ |\langle l | L_z | \mathcal{K} \rangle|^2 / (E_{\mathcal{K}} - E_l) ],$$

with  $N$  as Avogadro's number,  $L_z$  as the  $z$  component of orbital angular momentum which couples the ground state  $|l\rangle$  with the excited states  $|\mathcal{K}\rangle$ , with energies  $E_l$  and  $E_{\mathcal{K}}$ , respectively. For NiO,  $E_{\mathcal{K}} - E_l = 1.05$  eV as determined by optical measurements.<sup>10</sup> This yields  $\chi_{VV} = 3.3 \times 10^{-6}$   $\text{cm}^3/\text{g}$  ( $= 2.46 \times 10^{-4}$   $\text{cm}^3/\text{mole}$ ). The diamagnetic susceptibility  $\chi_d$  for NiO is estimated to be  $\approx -0.5 \times 10^{-6}$   $\text{cm}^3/\text{g}$ , yielding  $\chi_{VV} + \chi_d \approx 2.8 \times 10^{-6}$   $\text{cm}^3/\text{g}$  as the correction which should be applied to the measured  $\chi_{||}^*$  and  $\chi_p$ . Hutchings and Samuelson<sup>11</sup> have estimated  $\chi_{VV} + \chi_d = (3.0 \pm 0.8) \times 10^{-6}$   $\text{cm}^3/\text{g}$  for NiO. The left-hand scale of Fig. 2 represents the derived  $\chi_{||}$  and  $\chi_{\perp}$  after  $\chi_{||}^*$  and  $\chi_p$  have been corrected for  $\chi_{VV} + \chi_d = 3.2 \times 10^{-6}$   $\text{cm}^3/\text{g}$ . With this correction  $\chi_{||}$  approaches 0 as  $T \rightarrow 0$  K,  $\chi_{\perp}(0) \approx 6.8 \times 10^{-6}$   $\text{cm}^3/\text{g}$ , and  $\chi(T_N) = 8.8 \times 10^{-6}$   $\text{cm}^3/\text{g}$ .

Next we consider the temperature dependence of  $\chi_{\perp}$  and  $\chi_{||}$ . Beginning with  $\chi_{\perp}(0)$ ,  $\chi_{\perp}$  remains constant to about 120 K and then it increases with increasing temperatures up to about  $T_N$  (Fig. 2). There is no broad minimum in  $\chi_{\perp}$  between 0 K and  $T_N$  as, for example, observed in MnO,<sup>4</sup> CoO,<sup>5</sup> RbMnF<sub>3</sub>,<sup>12</sup> and MnF<sub>2</sub>.<sup>13</sup> In the torque measurements of Kurosawa *et al.*,<sup>6</sup> a temperature-independent  $\chi_{\perp} \approx 9.1 \times 10^{-6}$   $\text{cm}^3/\text{g}$  was inferred from the data of  $\Delta\chi = \chi_{\perp} - \chi_{||}$  down to 77 K. Although this estimate is close to our magnitude of  $\chi_{\perp}(T_N)$ , our measurements show that  $\chi_{\perp}$  has significant temperature dependence as the temperature is lowered toward 0 K. The observed difference between  $\chi_{\perp}(0)$  and  $\chi_{\perp}(T_N)$  in our measurements is consistent with the theoretical expectations as explained below.

On the basis of spin-wave theory,<sup>14</sup> the relationship between  $\chi_{\perp}(0)$  and  $\chi_{\perp}(T_N)$  is given by

$$\chi_{\perp}(0) = \frac{\chi_{\perp}(T_N)}{(1 + \alpha/2)} \left[ 1 - \frac{\Delta S}{S} - \frac{e(\alpha)}{(2 + \alpha)ZS} \right]. \quad (1)$$

In Eq. (1),  $\alpha = H_A/H_E$  (anisotropy field/exchange field),

$\Delta S$  is the zero-point spin reduction,  $Z$  is the number of neighbors with which exchange coupling is dominant,  $e(\alpha)=0.582$  for  $\alpha < 10^{-2}$ . For NiO,  $\alpha$  can be determined from the out-of-plane anisotropy parameter  $D_1$  and next-nearest-neighbor exchange  $J_2$  (which determines  $T_N$  and is dominant in NiO). One can estimate  $D_1$  from  $\chi_{\perp}(0)$  and the upper mode  $\omega_1$  of antiferromagnetic resonance using the relation<sup>11</sup>

$$(\hbar\omega_1)^2 = Ng^2\mu_B^2 S^2 D_1 / \chi_{\perp}(0), \quad (2)$$

where  $N$  is the number of  $\text{Ni}^{2+}$  ions/g. Using  $\hbar\omega_1=52.7$  K,  $g=2.23$ ,  $\chi_{\perp}(0)=6.8 \times 10^{-6}$  cm<sup>3</sup>/g, and  $S=1$ , we get  $D_1=0.77$  K. This estimate differs from the value of  $D_1=1.13$  K determined from the fit of the spin-wave dispersion curves.<sup>9</sup> As we note later, the experimental value of  $S=0.81$  at 4.2 K (Ref. 15) and this value in Eq. (2) gives  $D_1=1.15$  K, in excellent agreement with the magnitude of  $D_1$  determined in Ref. 11.

Using  $D_1=1.15$  K and  $J_2=202$  K as determined later in this paper,  $\alpha=0.0057$ . Thus  $e(\alpha)\simeq e(0)=0.582$  in Eq. (1). For cubic systems, the spin-wave theory gives  $\Delta S/S=0.078/S$  which in turn gives  $\Delta S=0.078$  for  $S=1$  for  $\text{Ni}^{2+}$ . Using these estimates and  $\chi_{\perp}(T_N)=8.8 \times 10^{-6}$  cm<sup>3</sup>/g in Eq. (1), we get  $\chi_{\perp}(0)=7.7 \times 10^{-6}$  cm<sup>3</sup>/g, to be compared with the experimental value of  $6.8 \times 10^{-6}$  cm<sup>3</sup>/g. If we use  $\Delta S=0.19$  for  $\text{Ni}^{2+}$  as determined by the neutron-scattering experiments of Alperin<sup>15</sup> at 4.2 K, then Eq. (1) gives  $\chi_{\perp}(0)=6.7 \times 10^{-6}$  cm<sup>3</sup>/g, in excellent agreement with the experimental value. Thus for NiO, the experimental value of  $\chi_{\perp}(0)$  is consistent with a large magnitude of spin reduction ( $\Delta S \simeq 0.19$ ) for  $\text{Ni}^{2+}$  observed in neutron-scattering experiments.<sup>15</sup>

Next the temperature dependence of  $\chi_{\parallel}$  is examined. Including the effect of anisotropy, which introduces a gap  $kT_{AE}$  in the spin-wave spectrum, the temperature dependence of  $\chi_{\parallel}$  is given by<sup>13,14</sup>

$$\chi_{\parallel} = AT^2 f(T/T_{AE}), \quad (3)$$

where the constant

$$A = \frac{1}{6} Ng^2 \mu_B^2 k^2 (Z |J| S/2)^{-3}$$

and the function  $f(T/T_{AE})$  goes exponentially to 0 as  $T \rightarrow 0$  K and it approaches unity for  $T \gg T_{AE}$ . As noted earlier  $T_{AE}=52.7$  K for NiO.<sup>11</sup> We have fitted our data with Eq. (3) first by putting  $f=1$  and leading to  $A \simeq 2 \times 10^{-11}$  cm<sup>3</sup>/g deg<sup>2</sup>. This fit is shown as triangles in Fig. 3 where the solid line represents the experimentally derived value of  $\chi_{\parallel}$ . Note that  $f=1$  corresponds to neglecting the effect of anisotropy. Next, using the values of  $f(T/T_{AE})$  up to  $T/T_{AE}=5$  from Ref. 14, the data were also fitted to Eq. (3) and the fit in Fig. 3 (solid circles) corresponds to  $A \simeq 3 \times 10^{-11}$  cm<sup>3</sup>/g deg<sup>2</sup>. The magnitude of  $A$  is estimated to be accurate to about 20%. Deviations from spin-wave theory at higher temperatures are expected because of interactions among spin waves.

Using  $A=3 \times 10^{-11}$  cm<sup>3</sup>/g deg<sup>2</sup> determined above and the expression for  $A$ , one can estimate effective  $|J|Z=6(2J_1+J_2)$  in NiO where  $J_1$  is the nearest-neighbor exchange constant. With  $g=2.23$ , and  $S=0.81$  [experimental value of  $S=0.81$  at 4.2 K is consistent with

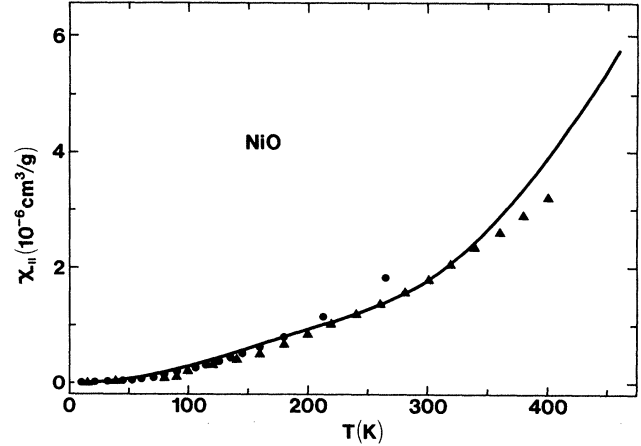


FIG. 3. Comparison of the derived  $\chi_{\parallel}$  (solid line) with Eq. (3). Solid circles represent Eq. (3) and triangles represent a special case of Eq. (3) with  $f=1$ , with the magnitude of  $A$  given in the text.

our  $\chi_{\perp}(0)$  and it also gives consistent value of  $D_1$  as noted earlier], we get  $2J_1+J_2=213$  K with about 20% accuracy, this being determined by the accuracy of  $A$ . In the next section we determine  $J_1=34$  K and  $J_2=202$  K with about 10% uncertainty. Thus  $2J_1+J_2=213$  K determined from the fit to Eq. (3) is consistent with these values of  $J_1$  and  $J_2$ , considering the uncertainties involved in these determinations.

Near  $T_N$ , the temperature variation of magnetic susceptibilities can determine the nature of phase transition at  $T_N$ . From the behavior of  $\partial(\chi_p T)/\partial T$  near  $T_N$ , the nature of magnetic transitions in NiO and in MnO, Fe<sub>2</sub>O, and CoO has been discussed in detail in a recent paper.<sup>16</sup> Here we just note that using the data on both the stressed-crystal and powder sample, the transition in NiO has been found to be of second order.<sup>16</sup>

#### IV. EXCHANGE CONSTANTS

Using the experimental susceptibilities reported here, and after correcting for  $\chi_{VV}$  and  $\chi_d$ , the magnitudes of exchange constants of NiO are now determined. For type-2 ordering of NiO, with  $J_1$  and  $J_2$  as the nearest- and next-nearest-neighbor exchange constants, respectively, the molecular-field approximation gives<sup>17</sup>

$$T_N = 2J_2 S(S+1), \quad (4)$$

$$\chi(T_N) = Ng^2 \mu_B^2 / 12(J_1 + J_2). \quad (5)$$

Using  $\chi(T_N)=8.8 \times 10^{-6}$  cm<sup>3</sup>/g ( $=6.57 \times 10^{-4}$  cm<sup>3</sup>/mole) from Fig. 1,  $S=1$ , and  $g=2.23$ , we get  $J_1=105$  K and  $J_2=131$  K for  $T_N=524$  K. (Near  $T_N$ , the use of  $S=1$  is reasonable since zero-point spin reduction is only applicable well below  $T_N$ .) It is well known that the molecular-field approximation usually does not give good quantitative estimates. Therefore, we attempt estimates of  $J_1$  and  $J_2$  using the random-phase-approximation Green's-function theory.<sup>17</sup> In this theory the expression for  $\chi(T_N)$  is the same as Eq. (5) whereas  $kT_N/J_2$  is a function of  $S$  and the ratio  $J_1/J_2$ . Bartel

and Morosin<sup>18</sup> have calculated  $kT_N/J_2$  versus  $J_1/J_2$  for  $S=1$ . From Eq. (5), one can calculate  $(J_1+J_2)$  which for NiO with  $g=2.23$  gives  $(J_1+J_2)=236$  K for  $\chi(T_N)=8.80\times 10^{-6}$  cm<sup>3</sup>/g ( $=6.57\times 10^{-4}$  cm<sup>3</sup>/mole). Now to get  $J_1$  and  $J_2$ , we use an iterative procedure and Fig. 1 of Ref. 18, ( $kT_N/J_2$  versus  $J_1/J_2$ ). We assume  $J_1=0$  which corresponds to  $kT_N/J_2=2.67$  yielding  $J_2=196$  K for  $T_N=524$  K. From  $(J_1+J_2)=236$  K, and  $J_2=196$  K,  $J_1=40$  K is obtained. This in turn yields  $J_1/J_2=0.20$ . Again using Fig. 1 of Ref. 18, this value of  $J_1/J_2$  corresponds to  $kT_N/J_2=2.60$ , yielding  $J_2=202$  K. Again using  $J_1+J_2=236$  K, this leads to new  $J_1=34$  K and  $J_1/J_2=0.17$ . Again Fig. 1 of Ref. 18 and  $J_1/J_2=0.17$  yield  $kT_N/J_2=2.60$ , the same value as before. Thus this rapid convergence assures us that  $J_1=34$  K and  $J_2=202$  K are the best estimates that can be obtained with the use of the random-phase-approximation Green's-function theory. The above estimates of  $J_1$  and  $J_2$  are not too different from those determined by Hutchings and Samuelsen<sup>11</sup> from the fit to the experimental spin-wave dispersion curves, for a polydomain sample. They reported  $J_2=221\pm 4$  K and  $J_1=-16.0^{+10}_-6$  K whereas from the Raman scattering data, Dietz *et al.*<sup>19</sup> reported  $J_2=230$  K assuming  $J_1$  to be near 0. One reservation about the estimates of  $J_1$  and  $J_2$  determined from spin-wave dispersion curves is that the measurements were not carried out in a single- $T$ -domain crystal.

A major contribution of this work is that the accurate

values of magnetic susceptibilities of NiO at various temperatures are now known. The accuracy of the exchange constants determined from these susceptibilities is determined by the accuracy of the theories. The facts are that  $J_1$  and  $J_2$  determined from the molecular-field approximation ( $J_1=105$  K,  $J_2=131$  K) are vastly different from their magnitudes determined from the Green's-function theory ( $J_1=34$  K,  $J_2=202$  K), the latter, however, being in reasonable agreement with the values obtained from the spin-wave dispersion curves ( $J_1=-16$ ,  $J_2=221$  K) and from Raman scattering ( $J_1\approx 0$ ,  $J_2=230$  K). In MnO, the exchange constants determined from  $\chi$  values with the use of the Green's-function theory agreed well with those determined from the fit to the spin-wave dispersion curves.<sup>4,17,20</sup> If the same accuracy is adopted for  $J_1=34$  K and  $J_2=202$  K determined here for NiO, then the uncertainty in these magnitudes should not be more than 10%. Finally, the effect of exchange-striction on  $J_1$  and  $J_2$  values below  $T_N$  should be considerably smaller in NiO as compared to that in MnO (Refs. 17 and 18) because of its smaller magnitude in NiO.<sup>11</sup> It is for this reason that it has been ignored in the discussion presented in this work.

#### ACKNOWLEDGMENT

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<sup>1</sup>W. L. Roth, Phys. Rev. **110**, 1333 (1958); **111**, 772 (1958); J. Appl. Phys. **31**, 2000 (1960).

<sup>2</sup>H. La Blanchetais, J. Phys. Radium **12**, 765 (1951).

<sup>3</sup>J. R. Singer, Phys. Rev. **104**, 929 (1956).

<sup>4</sup>M. S. Jagadeesh and M. S. Seehra, Phys. Rev. B **23**, 1185 (1981).

<sup>5</sup>P. S. Silinsky and M. S. Seehra, Phys. Rev. B **24**, 419 (1981).

<sup>6</sup>K. Kurosawa, M. Miura, and S. Saito, J. Phys. C **13**, 1521 (1980).

<sup>7</sup>M. S. Seehra and M. S. Jagadeesh, Phys. Rev. B **20**, 3897 (1979).

<sup>8</sup>Magnetic field dependence of  $\chi_{\parallel}^*$  was also measured at 297 and 77 K in separate experiments. At 77 K with increasing fields,  $\chi_{\parallel}^*$  increases from  $6.8\times 10^{-6}$  cm<sup>3</sup>/g at 200 Oe to a saturating value of  $7.2\times 10^{-6}$  cm<sup>3</sup>/g above 2 kOe. On decreasing fields, hysteresis is observed below 2 kOe in that  $\chi_{\parallel}^*$  increases with decreasing fields reaching a value of about  $8.0\times 10^{-6}$  cm<sup>3</sup>/g at 200 Oe. Similar results were observed at 297 K except the initial magnitude of  $\chi_{\parallel}^*$  was  $7.4\times 10^{-6}$  cm<sup>3</sup>/g, the saturating value  $8.0\times 10^{-6}$  cm<sup>3</sup>/g, and the magnitude on reversal  $8.3\times 10^{-6}$  cm<sup>3</sup>/g. This hysteresis of  $\chi_{\parallel}^*$  is probably due to  $S$ -wall displacement [see Refs. 4 and 6 and H. Kondo and T. Takeda, J. Phys. Soc. Jpn. **19**, 2041 (1964)]. The argument of  $\chi_{\parallel}^*=(\chi_{\parallel}+\chi_{\perp})/2$  therefore applies only to the initial  $\chi_{\parallel}^*$  as measured here at 200 Oe (on a virgin sample or a sample heated to above  $T_N$ ).

<sup>9</sup>Direct measurements of  $\chi_{\perp}$  in NiO are possible by orienting the sample so that  $\vec{H}||[111]$ , the stressed direction which is also

the hard-magnetization direction. (For measurements of  $\chi_{\perp}$  in MnO, see Ref. 4.) Several attempts, all unsuccessful, to measure  $\chi_{\perp}$  in NiO this way were made. The primary experimental difficulty is the rather large size of the stress cell for this orientation relative to the available space since the cell must hang vertically in the hangdown tube of the balance without touching the sides. This could not be achieved in these experiments. The fact that  $\chi_{\perp}$  for NiO is nearly an order of magnitude smaller than that of MnO puts additional strain on the experimental accuracy.

<sup>10</sup>L. Messick, W. C. Walker, and R. Glosser, Phys. Rev. B **6**, 3941 (1972).

<sup>11</sup>M. T. Hutchings and E. J. Samuelsen, Phys. Rev. B **6**, 3447 (1972).

<sup>12</sup>G. Chaddha and M. S. Seehra, Solid State Commun. **44**, 1097 (1982).

<sup>13</sup>C. Trapp and J. W. Stout, Phys. Rev. Lett. **10**, 157 (1963).

<sup>14</sup>See the review by F. Keffer, in *Handbuch der Physik* (Springer, Berlin, 1966), Vol. 18, p. 1.

<sup>15</sup>H. A. Alperin, J. Phys. Soc. Jpn. Suppl. **17-B3**, 12 (1962).

<sup>16</sup>G. Srinivasan and M. S. Seehra, Phys. Rev. B **28**, 6542 (1983).

<sup>17</sup>M. E. Lines and E. D. Jones, Phys. Rev. **139**, A1304 (1965); and references therein.

<sup>18</sup>L. C. Bartel and B. Morosin, Phys. Rev. B **3**, 1039 (1971).

<sup>19</sup>R. E. Dietz, G. I. Parisot, and A. E. Meixner, Phys. Rev. B **4**, 2302 (1971).

<sup>20</sup>M. F. Collins, V. K. Tandon, and W. J. L. Buyers, Int. J. Magn. **4**, 17 (1973).