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Weak ferromagnetism and magnetic interactions in La_2NiO_4

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Abstract. Experimental evidence of weak ferromagnetic behaviour in the low-temperature tetragonal phase of stoichiometric La_2NiO_4 is given from both isothermal magnetization and magnetic susceptibility studies. A clear anomaly is observed in the magnetic susceptibility around the first-order structural phase transition at $T_{c1} = 80$ K. The symmetric and antisymmetric exchange interactions in La_2NiO_4 are evaluated within the scope of the mean-field approximation and compared to those reported in La_2CuO_4. In spite of the strong similarities in the overall magnetic behaviour of La_2NiO_4 and La_2CuO_4 oxides, severe quantitative differences exist in the magnetic interactions, which lead to a strong reduction in the two-dimensional character in La_2NiO_4. It is suggested that this different magnetic behaviour may be related to the absence of superconducting behaviour in the Ni oxides.

1. Introduction

In the last few years, considerable effort has been devoted to the understanding of the unusual magnetic properties of CuO-based high-T_c superconductors, mainly because long-range 3D antiferromagnetic ordering of the copper sublattice appears as soon as the carrier concentration is low enough to suppress the superconducting state [1, 2]. The most promising line of thought to face this problem seems at present to be the comprehension of strongly correlated 2D electronic systems. Any modelling in this direction is based on knowledge of the exchange interactions and thus it seems crucial to relate the magnetic behaviour of the 2D systems to the value of those magnetic interactions.

It has lately been proved experimentally that the magnetic properties of La_2CuO_4 may be essentially understood within the scope of the $S = 1/2$ 2D antiferromagnetic Heisenberg model [3], with very high in-plane exchange interaction ($J \simeq 0.16$ eV [4]), the main perturbation to this model coming from the antisymmetric exchange interaction D ($D \simeq 0.55$ meV), which allows the development of a weak ferromagnetic out-of-plane component that couples antiferromagnetically from plane to plane [5–8]. In this framework, the observed long-range 3D antiferromagnetic ordering of the copper sublattice ($T_N \simeq 320$ K [9]) is directly related to the existence of an interplane exchange

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interaction J', which turns out to be rather small ($J' \simeq 0.003$ meV [7, 8]). Then, the ratio $J'/J \simeq 10^{-5}$ gives us a quantitative estimation of the strong 2D magnetic character of the La_2CuO_4 oxide.

A parallel study of the magnetic properties and magnetic interactions in the La_2NiO_4-type oxides seems to be very appealing as long as the magnetic behaviour of this system displays strong similarities with the Cu counterpart, but no superconductivity has been clearly evidenced in this case on doping with holes, although some authors have claimed the occurrence of superconductivity in the nickelates [10].

The crystallographic structure of La_2NiO_4 is described above 770 K in the tetragonal space group $I4/mmm$, which is known as the T structure of the K_2NiF_4-type compounds. Below this temperature, a second-order structural phase transition takes place [11, 12], leading to a new structure that can be described in the orthorhombic space group *Bmab*. This structural phase transition is of the same kind as that observed in La_2CuO_4 at $T \simeq$ 530 K [13]. The orthorhombic distortion may be easily understood: if we assume (a_T, b_T) to be the in-plane cell axes of the $I4/mmm$ tetragonal space group, the NiO_6 octahedra rotate along the $a_O = a_T + b_T$ axis, where $(a_O, b_O) = (a_T + b_T, a_T - b_T)$ stand for the in-plane cell axes of the *Bmab* orthorhombic space group. Furthermore, Rodríguez-Carvajal *et al* [12] (see also [14]) report that, assuming that the tilt of the NiO_6 octahedra is nearly rigid, on going from 250 K to 120 K the tilt angle varies from 5.3° to 5.8°. Then, when this angle is greater than a critical value, the *Bmab* structure becomes unstable and an orthorhombic-to-tetragonal (*Bmab* to $P4_2/ncm$) first-order structural phase transition occurs at $T_{c1} \simeq 80$ K in order to rearrange the structure. In this low-temperature structural phase transition, the NiO_6 octahedra rotate along the a_T axis, similarly to what was observed by Axe *et al* [15] in $La_{2-x}Ba_xCuO_4$. No transition of this kind is observed in stoichiometric La_2CuO_4.

Referring to the magnetic structure, stoichiometric $La_2NiO_{4.00}$ presents 3D long-range antiferromagnetic ordering below $T_N = 330$ K [12], as has been demonstrated by neutron diffraction. The magnetic structure of the orthorhombic (*Bmab*) high-temperature phase is essentially the same as La_2CuO_4, except that the spin direction is parallel, instead of perpendicular, to the magnetic propagation vector $k = [1\,0\,0]$. In this sense, the magnetic structure of La_2NiO_4 is described by the g_x mode [12, 16] as long as the crystallographic structure remains orthorhombic. Consequently, the magnetic moments are aligned antiferromagnetically along the a_O axis, so that the rotation of the NiO_6 octahedra along this axis does not affect them. Below T_{c1}, the situation is more complex because the low-temperature tetragonal ($P4_2/nmc$) structure allows the existence of a ferromagnetic component along the c axis ($g_x + c_y f_z$ or $g_x c_y f_z$ magnetic modes) [12, 17], which is related to the rotation of the NiO_6 octahedra along the a_T axis. On the other hand, the magnetic structure of La_2CuO_4 is described in the $g_y a_z$ mode, thus indicating that the Cu^{2+} magnetic moments are aligned antiferromagnetically along the b_O axis, in such a way that the rotation of the CuO_6 octahedra along the a_O axis gives way to a weak out-of-plane component, which orders antiferromagnetically. This a_z ($a_z \simeq 0$) mode is clearly evidenced by the appearance of a metamagnetic-like field-induced transition observed in the isothermal magnetization curves [7, 8].

We present an experimental study of both the DC magnetic susceptibility and the isothermal magnetization curves (in magnetic fields up to 5 T) of an antiferromagnetic polycrystalline La_2NiO_4 sample, which demonstrates the formation of a weak ferromagnetic component in the magnetic structure of the low-temperature tetragonal (LTT) phase. From the knowledge of (i) the Néel temperature, (ii) the zero-temperature canting angle, (iii) the magnetic moment of Ni^{2+} ions and (iv) the intraplane exchange

interaction (as deduced from the spin-wave stiffness constant determined by means of inelastic neutron scattering [16]), we are able to obtain a consistent set of symmetric and antisymmetric exchange constants of La_2NiO_4. All the magnetic signatures of stoichiometric La_2NiO_4 may be closely compared to those of La_2CuO_4, which we are confident may help to elucidate the relevance of the magnetic interactions for the high-T_c superconducting mechanisms.

2. Experimental details

The sample used in this study was prepared by solid-state reaction of high-purity NiO and La_2O_3 oxides that were previously dried. The reaction temperature was 1450 °C and all this first step was carried out in air. Several intermediate grindings were performed until long-exposure Guinier x-ray diffraction patterns showed no detectable amount of the single oxides. The second processing step to obtain stoichiometric samples was a hydrogen flow reduction at 350 °C for several hours, which was previously monitored by means of thermogravimetric analysis in order to avoid an excess of the reduction process, which would have led to the appearance of metallic Ni. The final purity of the samples was confirmed by x-ray powder diffraction, chemical titration (showing no detectable amount of Ni^{3+} ions) and finally high-temperature hysteresis loop magnetic measurements, which can detect any residual ferromagnetic Ni at the 10^{-4}–10^{-5} level. In our sample we estimated that a maximum content of 0.03% (by weight) of metallic Ni existed. The isothermal magnetization curves (up to 5 T) and the DC magnetic susceptibility ($H = 1.5$ kOe) were measured with a SQUID magnetometer (quantum design). Unless otherwise stated, the magnetization curves were recorded in decreasing magnetic field to minimize domain effects.

3. Results

In figure 1 we present the DC magnetic susceptibility measured with an applied field of 1.5 kOe. A marked anomaly appears between 50 and 95 K, with a sharp peak around $T \simeq 70$ K. Previous neutron powder diffraction (NPD) measurements [11] had signalled that the low-temperature orthorhombic-to-tetragonal first-order structural phase transition occurs at $T_{c1} \simeq 80$ K. Later Rodríguez-Carvajal *et al* [12] reported that high-resolution neutron powder diffraction showed that the tetragonal phase began to nucleate around 95 K and grew progressively on cooling, at the expense of the orthorhombic phase, and that because of its first-order character there was phase coexistence down to about 50 K. Those authors found that, at $T_{c1} \simeq 80$ K, the sample fraction of each structural phase was about 50%.

It is also obvious that down to 150 K the magnetic susceptibility decreases, matching the antiferromagnetic ordering at $T_N \simeq 330$ K [12, 17]. Nevertheless, below 150 K, the magnetic susceptibility up-turns and increases strongly, which is tentatively attributed to oxygen inhomogeneities, which would lead to clusters of paramagnetic Ni^{2+} (magnetic defects). Owing to the fact that below 100 K both effects (anomalous increase in susceptibility and structural phase transition) are superimposed, the inverse of the DC magnetic susceptibility does not display Curie–Weiss behaviour over a wide temperature range. We estimate the amount of paramagnetic Ni^{2+} ions in our sample to be less than 4% of the total amount of Ni^{2+} magnetic moments.

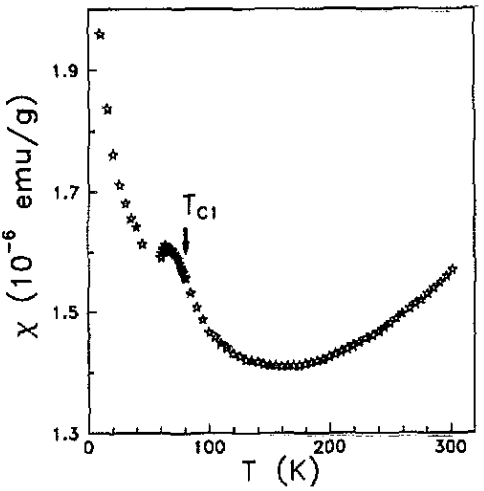

Figure 1. Low-field DC magnetic susceptibility ($H = 1.5$ kOe) of stoichiometric La_2NiO_4. $T_{c1} \simeq 80$ K signals the structural phase transition temperature determined by neutron powder diffraction.

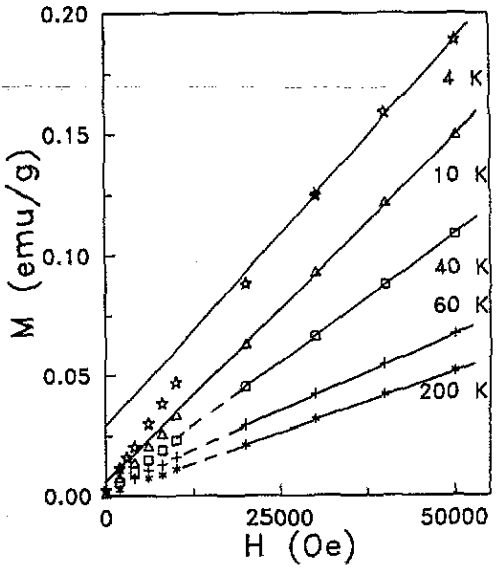

Figure 2. Isothermal magnetization curves $M(H)$ ($H_{max} = 5$ T) as a function of temperature.

Several typical isothermal magnetization curves are shown in figure 2, where the whole temperature range investigated (4–300 K) is displayed. Even a simple visual inspection makes it clear that a ferromagnetic component appears at low temperature. This is further corroborated by the low-field region of the hysteresis loop at $T = 4.2$ K, where the existence of some remanence ($M_r \simeq 10^{-3}$ emu g^{-1}) and a moderate coercive field of $H_c \simeq 700$ Oe is obtained, thus indicating a small anisotropy for the weak ferromagnetic spin component. Both the remanence and the coercive field are zero above $T_{c1} \simeq 80$ K.

All the experimental $M(H)$ curves may be represented, at high enough magnetic fields, by the equation $M(H) = M_0 + \chi_d H$ where both the zero-field extrapolated magnetization M_0 and the high-field magnetic susceptibility χ_d are strongly temperature-dependent. The results of our fitting procedure are displayed in figures 3(*a*) and (*b*).

Above 80 K and up to room temperature, the saturation magnetization is not exactly zero. We attribute this fact to the presence of a tiny amount of residual metallic Ni in our sample, which is a consequence of a slight excess in the reduction process. In the represented values of M_0 (figure 3(*a*)), we have deduced the contribution of this impurity, which we evaluate to be present in our sample at a concentration less than 0.03% by weight. Below 10 K, M_0 increases extraordinarily, and when extrapolating our experimental data to zero temperature we obtain $M_0(0) = 0.058$ emu g^{-1}, corresponding to $M_0(0) = 4.2 \times 10^{-3}\ \mu_B$/formula unit. It is obvious that this important low-temperature increase has nothing to do with the tiny amount of residual metallic Ni that is present in our sample. If we assume that the Ni^{2+} magnetic moments in La_2NiO_4 are $\mu = 1.68\ \mu_B$ [12], we obtain a canting angle $\theta = 0.14°$, which should be comparable to that reported for La_2CuO_4 ($\theta \simeq 0.17°$, weak ferromagnetic component at zero temperature 3×10^{-3}

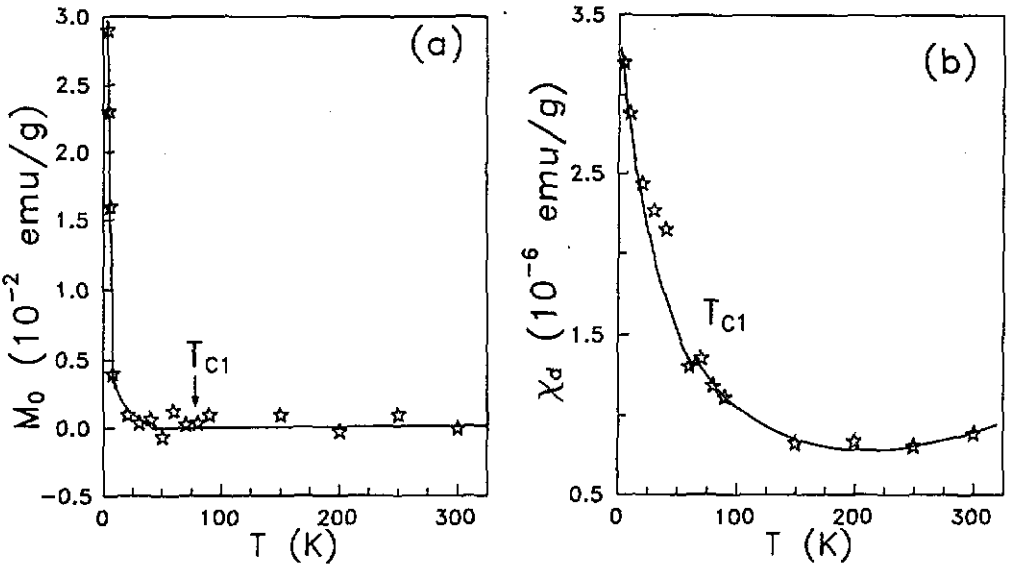

Figure 3. (*a*) Saturation magnetization M_0 and (*b*) high-field differential susceptibility χ_d as functions of temperature. $T_{c1} = 80$ K signals the structural phase transition temperature determined by neutron powder diffraction.

μ_B/FU [8]). Although we are not able to give a detailed explanation for this rapid increase in M_0, it might be related to some kind of spin reorientation leading to an increase in the magnetic interaction responsible for the weak ferromagnetic behaviour. We will come back to this point later on.

The high-field differential susceptibility χ_d (figure 3(*b*)) displays the same features as the DC magnetic susceptibility (figure 1): (i) a neat peak (above our resolution limit) at T_{c1}, which actually evidences the structural phase transition; (ii) a large increase at low temperature, probably signalling the existence of magnetic defects usually observed in this kind of antiferromagnetic oxides [18]; and (iii) a smooth decrease from room temperature down to 150 K, matching the antiferromagnetic ordering of the Ni^{2+} magnetic moments at $T_{N1} = 330$ K.

We should stress that the observed weak ferromagnetic component $M_0(T)$ is independent of the maximum applied field in our experiment, thus indicating that the magnetic anisotropy is small (as somehow the small coercive field also signals). This also suggests that the isotropic Heisenberg model should be an adequate first-order approximation, similarly to K_2NiF_4, for which it was recently shown by Birgeneau *et al* [19] that the experimental temperature dependence of the instantaneous antiferromagnetic spin–spin correlation length may be well represented by the theoretical $S = 1$ Heisenberg model proposed by Chakravarty *et al* [20]. It is also worth noting that we did not observe any field-induced transition in La_2NiO_4, in contrast to La_2CuO_4 [7, 8], but we were able to identify all the domain features typical of ferromagnetic systems (hysteresis loops, magnetic remanence, coercive field). This gives further support to the group-theoretical analysis [11, 12], which shows that the magnetic basis of the LTT phase of La_2NiO_4 is either $g_x + c_y f_z$ or $g_x c_y f_z$, contrary to La_2CuO_4, which is $g_y a_z$.

On the other hand, assuming $g_x + c_y$ and $g_x c_y$ to be the two in-plane magnetic modes allowed by the crystallographic symmetry below T_{c1}, NPD is unable to distinguish between

them since the low-temperature phase is tetragonal. In this sense, a hypothetical spin reorientation from the [1 0 0] direction to the [1 1 0] direction (or vice versa) would not be detected. However, this reorientation might have some effect on the ferromagnetic component (f_z mode), owing to the fact that, in both magnetic modes, $g_x + c_y f_z$ and $g_x c_y f_z$, the relative arrangement of the Ni^{2+} magnetic moments with respect to the tilt axis of the NiO_6 octahedra is different. Otherwise, although the origin of this phenomenon is not clear, it might be related to a change in the local easy magnetization axis associated with the magnetocrystalline anisotropy of Ni ions, leading to the mentioned up-turn of the saturation magnetization at about 10 K (figure 3(*a*)). It should be stressed that the same kind of in-plane spin reorientation is well-established in Nd_2CuO_4 [21].

4. Discussion

Let us now evaluate which are the main magnetic interactions among Ni ions in La_2NiO_4 in the light of the available experimental investigations and compare these results with those obtained in La_2CuO_4.

It has been experimentally shown that the 2D antiferromagnetic (AF) Heisenberg model is able to describe the in-plane instantaneous spin–spin correlations in La_2CuO_4 [3] and K_2NiF_4 [19], although some uncertainty still exists about the mechanism that drives the transition to the 3D long-range ordering. Some authors suggest that the 2D in-plane Ising anisotropy could be responsible for this fact, as seems to happen, for example, in K_2NiF_4 [19], while others point out that the 3D ordering is caused by the interplane magnetic interaction, as seems to happen, for example, in La_2CuO_4 [7, 8].

Let us first assume that La_2NiO_4 and La_2CuO_4 are good examples of the 2D antiferromagnetic Heisenberg model. In this basic framework, the Hamiltonian describing the magnetic interactions in either a NiO_2 or CuO_2 single plane may be written

$$\mathcal{H} = \sum_{NN} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j \tag{1}$$

where J_{ij} is the antiferromagnetic in-plane exchange interaction and the sum is over nearest neighbours. From this Hamiltonian, Chakravarty, Halperin and Nelson have developed a quantitative theory (CHN theory) [20] to account for the instantaneous spin–spin antiferromagnetic in-plane correlation length, which may be written as

$$\begin{aligned} S = 1/2 \qquad & l/a = C_1 \exp(2\pi\rho_S/k_B T) \\ S = 1 \qquad & l/a = C_2 \exp(2\pi\rho_S/k_B T)/(1 + k_B T/2\pi\rho_S) \end{aligned} \tag{2}$$

where a is the lattice parameter, C_1 and C_2 are constants and ρ_S is the spin stiffness constant,

$$2\pi\rho_S = 2\pi J S^2 (1 + 0.158/2S)^2 (1 - 0.552/2S) \tag{3}$$

thus implying that the analysis of any experimental result within the scope of this model requires knowledge of the intraplane superexchange interaction J.

For La_2CuO_4, further support for the assumption of 2D behaviour has recently been provided by Endoh *et al* and Yamada *et al* (see [3]), since the experimental results of the spin correlation length obtained from inelastic neutron scattering may be very nicely fitted to the formula proposed in the CHN theory, assuming the intraplane interaction obtained from neutron scattering ($J \simeq 0.16$ eV) [4].

Referring to stoichiometric La_2NiO_4, there are no reports in the literature of a detailed experimental analysis of the temperature dependence of the spin correlation length, although there are some data for certain fixed temperatures. For example, Aeppli *et al* [16] report that, in a slightly oxidized sample with Néel temperature around $T_N \simeq 70$ K, the correlation length is greater than 50 Å at 228 K, decreasing to roughly 10 Å when crossing the tetragonal-to-orthorhombic transition temperature ($T \simeq 240$ K). Further support for our assumption is given by the recent results reported by Birgeneau *et al* [19], where the old experimental data of K_2NiF_4 can also be nicely fitted to equation (2). If we assume equation (2) to be true when $S = 1$, we can easily derive the spin correlation length from the intraplane Ni–Ni superexchange antiferromagnetic interaction. This constant is not precisely known in La_2NiO_4, the only experimental report being that of Aeppli *et al* [16], which, by means of inelastic neutron scattering, gives as a lower limit $J \simeq 0.02$ eV. We have also determined this value of J from the oxidized sample $La_2NiO_{4+\delta}$ [22]. In this sample, the long-range antiferromagnetic ordering has been suppressed by the holes created by the oxygen excess and the magnetic susceptibility follows a Curie–Weiss-like behaviour above 200 K. The extrapolated Curie–Weiss temperature is negative, indicating antiferromagnetic correlations, and, within the scope of the mean-field approximation, we evaluate $J \simeq 0.016$ eV, in close agreement with the value obtained by Aeppli *et al* [16].

It is clear then that a strong reduction of the intraplane interaction J in La_2NiO_4, as compared to La_2CuO_4, will immediately lead to the reduction of the AF instantaneous spin–spin correlations. Anyway, it should be stated that, in both oxides, the spin correlation length persists well above the Néel temperature, as has been demonstrated experimentally by Aeppli *et al* [4, 16].

On the other hand, it is now well known that in La_2CuO_4 the most important deviation from the 2D $S = 1/2$ Heisenberg model is derived from the Dzyaloshinsky–Moriya antisymmetric interaction [23, 24]. The slight rotation of the CuO_6 octahedra in the orthorhombic phase leads to the appearance of an out-of-plane component of the Cu^{2+} magnetic moments, caused by the mentioned antisymmetric interaction among the spins. This component orders antiferromagnetically along the c axis (imposed by the $g_y a_z$ mode that describes the magnetic structure), which lies at the origin of the metamagnetic-like field-induced transition observed in the isothermal magnetization curves when the magnetic field is applied along this axis.

Of all the theoretical approaches aimed at determining the influence of the antisymmetric interaction on the magnetic structure of La_2CuO_4, the model developed by Thio *et al* [8] has been especially successful. These authors introduce the antisymmetric interaction in equation (1), and a characteristic mean-field treatment leads them to an excellent quantitative fit of the magnetic susceptibility, which explains the sharp peak observed in the magnetic susceptibility at the Néel temperature [25], the in-plane gap in the magnon spectra [5] and the temperature dependence of the critical field [7, 8]. Let us assume that this model is essentially valid for La_2NiO_4 as well. The exchange Hamiltonian may now be written

$$\mathcal{H} = \sum_{NN} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \boldsymbol{D}_{ij} \cdot \boldsymbol{S}_i \times \boldsymbol{S}_j \tag{4}$$

where J_{ij} is the nearest-neighbour in-plane coupling constant and $\boldsymbol{D} = (0, D, 0)$ is the antisymmetric exchange interaction. The latter is expected to be of order $D \sim J\Delta g/g$ [24]. Then at $T = 0$ K, the antisymmetric interaction will cause the magnetic moments to stand at a canting angle θ with the basal plane that can be expressed as $\sin\theta = M_0(0)/$

$Ng\mu_B S$, where $M_0(0)$ accounts for the extrapolated zero-temperature out-of-plane component of the magnetization. By maximizing the energy gain in the ordered state, θ can also be written as $\tan(2\theta) = D/J$. Then, it is obvious that, assuming θ to be small, which is actually the case, the latter expressions lead to

$$\theta \simeq M_0(0)/Ng\mu_B S \tag{5}$$

$$\theta \simeq D/2J. \tag{6}$$

Thio *et al* [8] propose that the antisymmetric superexchange interaction D should be proportional to the overlap between the $d_{x^2-y^2}$ orbitals of neighbouring sites, which increases with the tilt angle φ of the CuO_6 octahedra (NiO_6 in our present case). Then, the antisymmetric interaction D may be rewritten $D \sim \varphi J \Delta g/g$, thus leading to an order-of-magnitude relationship between the canting angle of the magnetic moments θ and the tilt angle φ,

$$\theta \sim \varphi \Delta g/g. \tag{7}$$

In La_2NiO_4, we have experimentally found that $M_0(0) = 4.2 \times 10^{-3}\ \mu_B$/FU, which means that the zero-temperature canting angle is about $\theta \simeq 0.14°$ (equation (5)), where we have used $\mu(Ni^{2+}) = 1.68\ \mu_B$ (obtained from neutron powder diffraction [12]). This angle is practically the same as those derived above in La_2CuO_4 [7, 8]. Then, from equation (6) we obtain the antisymmetric exchange coupling $D \simeq 0.1$ meV and, assuming the typical $\Delta g/g \sim 0.16$ [26] and that the rotation of the octahedra is nearly rigid ($\varphi \simeq 4°$ [12]), equation (7) leads us to expect that the value of θ should be around 0.6°, which is in agreement with the order of magnitude estimated experimentally.

Concerning La_2CuO_4, Thio *et al* [8] derive $\theta = 0.17°$, while Cheong *et al* [7] report $\theta = 0.22°$, both from isothermal magnetization single-crystal curves. These results are in good agreement with $\theta \simeq 0.28°$, obtained from equation (7), using the typical $\Delta g/g \simeq 0.1$ found in $MgO:Cu^{2+}$ [8] and the reported rotation angle $\varphi \simeq 2.8$ [8]. Finally, Cheong *et al* [7] determine, from equation (6), $D \simeq 0.6$ meV, while Thio *et al* [8] use this equation to derive $\theta \simeq 14°$, assuming $D \simeq 0.55$ meV (from the in-plane gap in the magnon spectra [5]) and $J \simeq 116$ meV (from Raman scattering [27]). It is worth stressing that Thio *et al* [8] theoretically obtain from their mean-field treatment $J = 100$ meV, $D = 0.7(1)$ meV and $J' = 0.0013(1)$ meV.

Finally, let us assume that the 3D long-range AF ordering appears in La_2NiO_4 because of the weak interplane exchange interaction J' (as seems to happen in La_2CuO_4), although, up to now, no experimental result has allowed us to discriminate this mechanism from the 2D Ising anisotropy mechanism. It is worth noting that J' is a super-superexchange coupling, which cancels by symmetry at nearest neighbours when the structure is the tetragonal type K_2NiF_4. In this sense J' is somehow dependent on the orthorhombic distortion of the structure. On decreasing the temperature, the 2D AF correlations increase and, when the effective interplane interaction $[l(T)/a]^2 J'$ becomes comparable to the thermal energy $k_B T$, a 3D ordering occurs. Then, at the Néel temperature, we can write [16, 21]

$$(M_S/M_t)^2 [l(T_N)/a]^2 J' \simeq k_B T_N \tag{8}$$

where the factor M_S/M_t accounts for the relative importance of the zero-point quantum fluctuations [28], which are enhanced in the 2D Heisenberg antiferromagnetic systems. In La_2CuO_4, the $S = 1/2$ spin reduction is about 35% ($M_S \simeq 0.67\ \mu_B$, $M_t \simeq 1.1\ \mu_B$) [29], while in La_2NiO_4, the $S = 1$ spin reduction is about 15% ($M_S \simeq 1.68\ \mu_B$, $M_t = 2\ \mu_B$) [12].

In this way, as the Néel temperature is nearly the same in both oxides and the correlation length is much greater in La_2CuO_4 than in La_2NiO_4, the interplane interaction J' would be larger in the latter than in the former ($J'(\mathrm{Ni}) > J'(\mathrm{Cu})$). We estimate from equations (2), (3) and (8) that, in La_2CuO_4, $J' = 0.007$ meV, while Thio *et al* [8] experimentally evaluate $J' = 0.0026(30)$ meV from the isothermal magnetization curves. On the other hand, this analysis leads us to $J' \simeq 1.1$ meV in La_2NiO_4. Further, at the Néel temperature, $l(T_N)/a \simeq 135$ in La_2CuO_4, while $l(T_N)/a \simeq 6$ in La_2NiO_4, thus implying that the in-plane correlation length in the La_2NiO_4-type oxides is much smaller than in the La_2CuO_4 type.

At the same time, the ratio J'/J is a measure of the two-dimensionality of the magnetic structure. From our values, we obtain that $J'/J \simeq 2 \times 10^{-5}$ and 5×10^{-2} in La_2CuO_4 and La_2NiO_4, respectively. This result could tentatively indicate that when going from Cu to Ni we move away from a 2D system. The main contribution to this increase in the J'/J ratio comes from the relative increase in J'. This is easy to understand when we consider the important chemical-bond differences between the two systems. The interplane superexchange interaction J' proceeds through the apical oxygen O(2) of the NiO_6 and CuO_6 octahedra, and this distance is strongly modified when going from Cu to Ni (2.39 Å versus 2.24 Å), thus giving an *a posteriori* justification of the enhanced 2D behaviour of La_2CuO_4. The change in the intraplane distances is much less pronounced (1.91 Å versus 1.94 Å).

5. Conclusions

We have found a reduction of both the intraplane interaction J and the effective in-plane correlation length l/a in La_2NiO_4 as compared to La_2CuO_4. On the other hand, if we assume that the interplane magnetic interaction J' drives the transition to the 3D long-range magnetic ordering, we derive that J' is much greater in La_2NiO_4 than in La_2CuO_4. In this framework, we obtain that in La_2NiO_4 the most important deviation from the 2D antiferromagnetic Heisenberg model would come from the interplane interaction ($J \simeq 20$ meV $> J' \simeq 1.1$ meV $> D \simeq 0.1$ meV), in contrast to La_2CuO_4, where the most important deviation is caused by the antisymmetric interaction ($J \simeq 160$ meV $> D \simeq 0.55$ meV $> J' \simeq 0.003$ meV). It is obvious that further experimental evidence should be obtained in order to ascertain which is the origin of the 3D ordering in the nickelates.

In conclusion, from a magnetic point of view, although La_2NiO_4 and La_2CuO_4 are qualitatively similar, important quantitative differences remain, mostly in the 2D character, which is strongly reduced in the nickelates. It is clear then that, in addition to the expected differences in the electronic structure of Cu and Ni oxides, any theoretical model should consider that the 2D character usually assumed in all the models presently adopted to understand the occurrence of high-T_c superconductivity in Cu oxides may not be an accurate approximation in Ni oxides. Actually, it may be suggested that this decrease in the 2D magnetic behaviour of the nickelates may have something to do with the absence of superconductivity. Nevertheless, we should also stress that the existence of a first-order structural phase transition around 80 K in La_2NiO_4 leads to the appearance of some anomalies in the magnetic properties, which might be in some way related to the claim of superconducting behaviour around this temperature [10].

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