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The effect of excess oxygen on long-range magnetic order in $\text{La}_2\text{NiO}_{4+\delta}$

Yu G Pogorelov†

Departamento de Física Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid, Spain

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Abstract. In the framework of a continuum model based on the microscopic magnetic symmetry properties of the metal oxide compound $\text{La}_2\text{NiO}_{4+\delta}$, a theoretical analysis is made of the stability of the antiferromagnetic (AFM) long-range order in the presence of impurity centres of excess oxygens at $\delta \ll 1$. The resulting magnetic phase diagram in the variables δ and T is compared with the recent experimental data for this system. Also the similarities and dissimilarities between the structurally isomorphic systems $\text{La}_2\text{NiO}_{4+\delta}$ and $\text{La}_2\text{CuO}_{4+\delta}$ are traced and their possible connection with superconducting properties is mentioned.

1. Introduction

Considerable attention has been paid to the effects of various dopants on the magnetic properties of layered perovskite metal-oxide systems [1]. Generally, these effects are most striking for the simplest single-layered compounds such as La_2CuO_4 , the prototype compound for the family of high- T_c superconductors [2]. Here, as well as interest in the general physical properties of low-dimensional and disordered magnets [3], there is the possible close connection between magnetic and superconducting correlations in the metallic state of these compounds at higher levels of doping. In the latter case, a great deal of work has already been done, treating the effect of mobile carriers on AFM correlations [4]. However, the initial stage of the evolution of these systems with doping, corresponding to a purely insulating state when no itinerant mechanisms apply, has so far received much less attention.

Recent analysis [5] of La_2CuO_4 shows that the main impurity effect on long-range AFM order, a drastic fall of the Néel temperature T_N for even small amounts, $c \ll 1$, of alkaline-earth ions or superstoichiometric oxygens, is apparently due to the local perturbations of Cu^{2+} magnetic anisotropy resulting from static deformations of the crystalline lattice produced by impurity ions. At the same time, the effects of indirect interaction between the spins of localized carriers via magnon excitations can only be of importance at very low temperatures $T \ll T_N$, which possibly accounts for observable spin-glass-like properties in this region [6].

The systems La_2NiO_4 and La_2CuO_4 are closely related in many aspects, possessing the same orthorhombic $Cmca$ crystalline structure [7] with just the same values of

† On leave of absence from the Institute of Metal Physics, Academy of Science of Ukraine, 252142 Kiev, Ukraine.

lattice parameters, namely $a = 5.473 \text{ \AA}$ and 5.356 \AA , $c = 5.532 \text{ \AA}$ and 5.399 \AA , $b = 12.541 \text{ \AA}$ and 13.167 \AA , and tilting angles of CuO_6 octahedra along the $\langle 001 \rangle$ directions of $\psi = 5.4^\circ$ and 5° respectively [8]. However, there is a qualitative distinction between the magnetic structures of these compounds in their AFM ordered phase, although they are both characterized by the planar spin alignment. Namely, the ground-state orientation of Cu^{2+} spin S_{in} ($S = 1/2$) at the n th site of the i th magnetic sublattice in La_2CuO_4 is nearly parallel to $\langle 001 \rangle$, that is to the direction of displacements of apex oxygens in the n th site's surrounding CuO_6 octahedron. In contrast to this, the $\langle 100 \rangle$ orientation of Ni^{2+} spins ($S = 1$) in La_2NiO_4 is orthogonal to these displacements (figure 1).

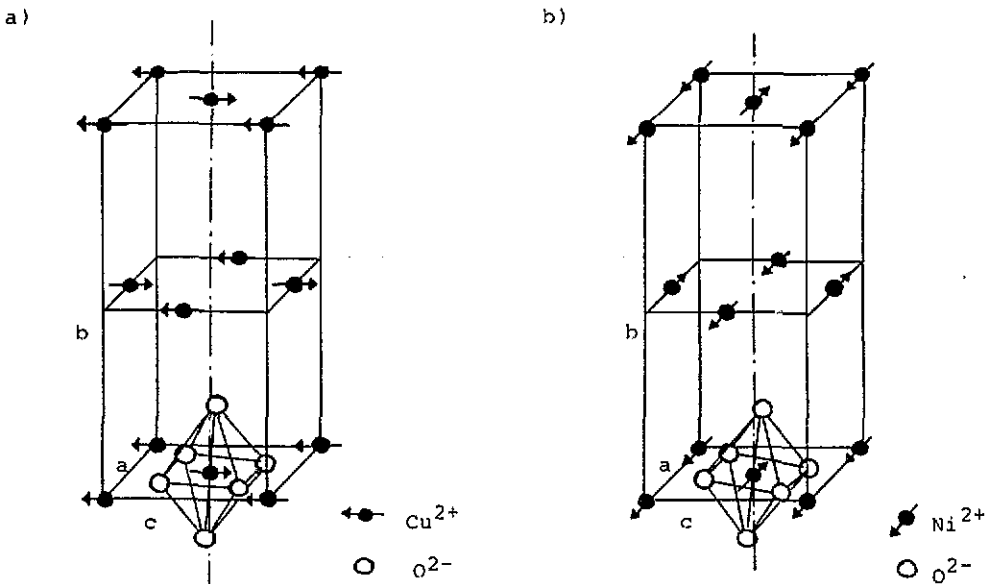


Figure 1. Magnetic configurations in the ground state for two structurally isomorphic systems: (a) La_2CuO_4 and (b) La_2NiO_4 .

The origin of this dissimilarity lies in the different nature of the spin anisotropies of the two cases. For Cu^{2+} spin $S = 1/2$ the anisotropy can come only from the Dzyaloshinskii–Moriya antisymmetric exchange interaction (AEI), $-d \cdot (S_{1n} \times S_{2n+\rho})$, between nearest neighbours from different sublattices (separated by the vector ρ), which is also confirmed by the presence of small out-of-plane canting $\simeq 0.17^\circ$ of Cu^{2+} spins and corresponding weak ferromagnetic moments of each CuO_2 plane in La_2CuO_4 [9]. For Ni^{2+} ($S = 1$), however, single-ion anisotropy (SIA), $-\lambda(S_z^2)^2$, is also possible and is more favourable; the ground-state magnetic symmetry eliminates AEI, providing a purely planar spin configuration for La_2NiO_4 [10].

The absence of visible dispersion of magnon excitations in the direction normal to the basal plane for both compounds [11, 12] indicates a quasi-two-dimensional character of spin interactions and suggests that the only stabilizing factor assuring a finite T_N value in both cases is the in-plane spin anisotropy λ , so that [13]

$$T_N \propto JS(S+1) \ln^{-1}(J/\lambda) \quad (1)$$

where J is the parameter of strong superexchange interaction between spins in the basal plane, and for the case of AEI $\lambda \simeq d^2/J$.

Remarkably, under doping, by superstoichiometric oxygens, for example, the two systems reveal a similarity in that their long-range AFM order is strongly affected for even small amounts, δ of a few percent, of dopants which is expressed in a considerable $T_N(\delta)$ dependence. But there are also qualitative dissimilarities in the character of these dependencies. In particular, in $La_2NiO_{4+\delta}$, unlike in $La_2CuO_{4+\delta}$ [14], no experimental evidence was found for an abrupt vanishing of T_N up to $\delta \simeq 0.1$ [10].

The aim of this paper is to develop a theoretical approach, similar to that of [5], for the $La_2NiO_{4+\delta}$ system and trace how the distinctions in the microscopic spin interactions for these two systems can reflect on their macroscopic magnetic properties. The theoretical results are compared with the available experimental data.

2. The microscopic Hamiltonian and phenomenological density of energy

We start with the microscopic spin Hamiltonian for a pure crystal of La_2NiO_4 , which being restricted to a single NiO_2 plane reads as

$$\mathcal{H}_0 = \sum_{n,\rho} [J S_{1n} S_{2n+\rho} - \Delta J S_{1n}^y S_{2n+\rho}^y] - \lambda \sum_{n,i} (S_{in}^z)^2. \quad (2)$$

Here the first sum describes the superexchange interaction between nearest neighbour Ni^{2+} spins, which is slightly anisotropic ($\Delta J \ll J$) because of small tetragonal distortions ($b/3 - a/\sqrt{2} \ll a$) of the ideal perovskite structure, and the form of which fully coincides with the corresponding term used in [5] for La_2CuO_4 . The SIA term in (2) is determined by the rhombic distortions of structure so, within an accuracy to the lowest order in ψ , we can put $\lambda \simeq q\psi^2$.

Performing a standard diagonalization procedure in (2) using the Holstein-Primakoff expansion and subsequent canonical u - v transformation, we arrive at the harmonic approximation to the magnon Hamiltonian:

$$\mathcal{H}_m = \sum_{j,k} \omega_{jk} \beta_{jk}^\dagger \beta_{jk} \quad (3)$$

where the eigen-frequencies of two branches of magnon excitations ($j = 1, 2$) are

$$\omega_{jk} = S \{ [(J + (-1)^j \Delta J \gamma_k / 2) z + 2\lambda]^2 - (J - \Delta J / 2)^2 z^2 \gamma_k^2 \}^{1/2} \quad (4)$$

$$\gamma_k = z^{-1} \sum_{\rho} \exp(ik \cdot \rho)$$

z being the number of nearest Ni^{2+} neighbours in the square NiO_2 lattice. The corresponding Bose operators relate to the spin operators as:

$$\beta_{1k} = \frac{1}{\sqrt{2SN}} \sum_n [u_{1k} (S_{1n}^+ + S_{2n}^+) + v_{1k} (S_{1n}^- + S_{2n}^-)] \exp(ik \cdot n)$$

$$\beta_{2k} = \frac{1}{\sqrt{2SN}} \sum_n [u_{2k} (S_{1n}^+ - S_{2n}^+) + v_{2k} (S_{1n}^- - S_{2n}^-)] \exp(ik \cdot n)$$

$$u_{jk} = \frac{1}{2} \sqrt{[(J + (-1)^j \Delta J \gamma_k / 2) z + 2\lambda] S / \omega_{jk} + 1} \quad v_{jk} = \sqrt{u_{jk}^2 - 1}.$$

From a comparison of (4) with the experimental data [10] for energy gap values $\omega_{1k=0} \simeq 4\sqrt{J\lambda} \simeq 3$ meV and $\omega_{2k=0} \simeq 4\sqrt{J(\lambda + 2\Delta J)} \simeq 7$ meV and magnon velocities (defined by $\omega_{jk} \simeq JS\sqrt{z}ak = sk$ at $k \gg \omega_{jk=0}/s$), $s \simeq 300$ meV Å, we can deduce the parameter values for the Hamiltonian (2) as

$$J \simeq 27.3 \text{ meV} \quad \Delta J \simeq 0.03 \text{ meV} \quad \lambda \simeq 0.02 \text{ meV}. \quad (5)$$

In what follows, in studying the influence of dopants upon system stability, we shall use, as in [5], a continual approximation, characterizing the crystal magnetic configuration by the two angles θ and φ which the staggered magnetization constitutes with the basal plane and the z -axis, respectively, both being continuous functions of the two-dimensional radius vector \mathbf{r} . Thus we proceed from (2) to the classical energy density

$$\begin{aligned} \mathcal{E}_0[\theta(\mathbf{r}), \varphi(\mathbf{r})] = S^2 z \{ & \frac{1}{2} J \cos^2 \theta [\nabla \varphi(\mathbf{r})]^2 + [J - \frac{1}{2} \Delta J \sin^2 \theta(\mathbf{r})] [\nabla \theta(\mathbf{r})]^2 \} \\ & + (S/a)^2 [\Delta J z \sin^2 \theta(\mathbf{r}) - 2\lambda \cos^2 \theta(\mathbf{r}) \cos^2 \varphi(\mathbf{r})]. \end{aligned} \quad (6)$$

Varying expression (6) with respect to $\theta(\mathbf{r})$ and $\varphi(\mathbf{r})$ we obtain two equations for the equilibrium distributions of the latter, the first of which readily gives $\theta(\mathbf{r}) = 0$ and, for $\varphi(\mathbf{r})$,

$$\nabla^2 \varphi(\mathbf{r}) - (1/2\xi_0^2) \sin 2\varphi(\mathbf{r}) = 0 \quad \xi_0 = a\sqrt{J/\lambda} \gg a \quad (7)$$

also giving $\varphi(\mathbf{r}) = 0$ for the ground-state configuration. Considering small fluctuations ($|\varphi| \ll 1$) over this ground state we can linearize (7), giving

$$\nabla^2 \varphi(\mathbf{r}) - \xi_0^{-2} \varphi(\mathbf{r}) = 0 \quad (8)$$

and obtain the simplest circular symmetric solution

$$\varphi(r) = \alpha K_0(r/\xi_0). \quad (9)$$

Here $K_0(x)$ is the zeroth-order MacDonald's function with asymptotics $K_0(x) \simeq -\ln(\gamma x/2)$, $\gamma = 1.781\dots$, $x \ll 1$, and $K_0(x) \simeq \sqrt{\pi/2x} e^{-x}$, $x \gg 1$. ξ_0 is seen to be the characteristic fluctuation length in the pure crystal and the constant α can be found by matching (9) to certain boundary conditions (as will be done below).

Since the anisotropy energy in (6) is invariant under the change $\varphi(\mathbf{r}) \rightarrow \varphi(\mathbf{r}) + \pi$, the minimum thermic activation energy E_{ac} necessary to turn over the magnetization against the ground state is provided by the formation of a magnetic fluctuation, satisfying (7), such that $\varphi = \pi$ in its centre, and decaying approximately as $[\pi/K_0(a/\xi_0)]K_0(r/\xi_0)$ at distances $r \gg a$ from the centre (whereas $\theta(\mathbf{r}) \equiv 0$ still). Inserting this distribution into (6) we get the approximation

$$E_{ac}^{(0)} \simeq \pi^2 J \ln^{-1}(J/\lambda) \quad (10)$$

that is of the order of (1). We can use the same approximation to evaluate the Néel temperature in the crystal containing impurities. Thus our aim is also to investigate how the ground-state configuration and activation energy will change after introducing dopants into the system (2).

As testified by several neutron scattering experiments, the superstoichiometric oxygens in La_2NiO_4 (as well as in La_2CuO_4) nearly occupy the $(1/4, 1/4, 1/4)$ sites (and their equivalent ones, according to the lattice symmetry), producing considerable static deformations of the crystalline lattice in their vicinity so the CuO_6 octahedra adjacent to the impurity site are tilted by $\psi_0 \simeq 20^\circ \gg \psi$ along the (101) directions [7]. The impurity contribution to the spin Hamiltonian can be represented by

$$\mathcal{H}_1 = -\lambda_0 \sum_{\mathbf{p}, \nu} (S_{j_{\mathbf{p}+\nu}}^x + \sigma(\mathbf{p}) S_{j_{\mathbf{p}+\nu}}^z)^2$$

where $\lambda_0 \simeq q\psi_0^2$, \mathbf{p} is a random impurity site occupied with the probability $c = \delta/2$, the vector ν runs over all Ni^{2+} spins within the area of inhomogeneous octahedra tilt around \mathbf{p} and $\sigma(\mathbf{p})$ is a random variable taking the values ± 1 , with equal probability, depending on the position of \mathbf{p} within the NiO_2 unit cell. After reading the continual limit, this gives the corresponding contribution to the energy density as

$$\mathcal{E}_1[\theta(\mathbf{r}), \varphi(\mathbf{r})] = -2\Lambda(S/a)^2 \sum_{\mathbf{p}} \cos^2 \theta(\mathbf{r}) \cos^2[\varphi(\mathbf{r}) + \sigma(\mathbf{p})\pi/4] \delta(\mathbf{r} - \mathbf{p}) \quad (11)$$

where $\Lambda = n_{\text{eff}}\lambda_0$, n_{eff} being the effective number of Ni^{2+} spins affected by a single impurity centre (provided the impurity concentration obeys the condition $c \ll n_{\text{eff}}^{-1}$ so different impurity centres do not overlap in the mean). In what follows we shall consider the ratio $\Lambda/\lambda = n_{\text{eff}}(\varphi_0/\varphi)^2 \gg 1$ as an independent parameter of impurity perturbation in the crystal.

3. Magnetic configurations in a crystal with impurities

In the presence of impurities in a crystal, variation of the total energy density

$$\mathcal{E}[\theta(\mathbf{r}), \varphi(\mathbf{r})] = \mathcal{E}_0[\theta(\mathbf{r}), \varphi(\mathbf{r})] + \mathcal{E}_1[\theta(\mathbf{r}), \varphi(\mathbf{r})]$$

with respect to $\theta(\mathbf{r})$ again gives $\theta(\mathbf{r}) = 0$ for the ground-state configuration, and then variation by $\varphi(\mathbf{r})$ results in

$$\nabla^2 \varphi(\mathbf{r}) - \frac{1}{2\xi^2} \sin 2\varphi(\mathbf{r}) - \frac{2\Lambda}{Jz a^2} \sum_{\mathbf{p}} \sin 2[\varphi(\mathbf{r}) + \sigma(\mathbf{p})\pi/4] \delta(\mathbf{r} - \mathbf{p}) = 0 \quad (12)$$

which generalizes (7) for this case. It is worth noting that the only essential distinction of (12) from the corresponding impurity contribution to the in-plane spin anisotropy of $\text{La}_2\text{CuO}_{4+\delta}$ [5] consists in the doubling of the sine argument. Hence there arise two possible directions of minimum energy near each \mathbf{p} in $\text{La}_2\text{NiO}_{4+\delta}$, as compared with the single direction in $\text{La}_2\text{CuO}_{4+\delta}$. This distinction of purely quantum nature between $S = 1/2$ and $S = 1$ requires a rather detailed analysis to be performed when studying the macroscopic behaviour of the system under doping.

Clearly, for finite c the uniform distribution $\varphi(\mathbf{r}) = 0$ cannot be a solution to (12) and the system ground-state configuration is, in general, a randomly fluctuating function, corresponding to chaotic distributions of \mathbf{p} , $\sigma(\mathbf{p})$. We begin its analysis

from the simplest case of a single impurity at $p = 0$ with $\sigma(p) = \sigma$. Supposing the fluctuation to be small even at the centre, that is

$$\varphi(0) \simeq \varphi(a) = \varphi_0 \ll 1 \quad (13)$$

we apply (9) with $\alpha \simeq \varphi_0 K_0^{-1}(a/\xi)$. Correspondingly, we obtain the impurity contribution to the ground-state energy

$$\begin{aligned} \Delta \mathcal{E} = & \int \{ \mathcal{E}[0, \varphi(r)] - \mathcal{E}_0[0, 0] \} d\mathbf{r} \simeq 2\pi S^2 \\ & \times \int_a^\infty \left\{ \frac{1}{2} J z [\nabla \varphi(r)]^2 + 2\lambda a^{-2} \sin^2 \varphi(r) \right\} d\mathbf{r} \\ & - 2\Lambda S^2 \cos^2(\varphi_0 + \sigma\pi/4) \simeq 8\pi J \varphi_0^2 \ln^{-1}(J/\lambda) - \Lambda(1 + \sigma \sin 2\varphi_0) \end{aligned}$$

and, minimizing this expression with respect to φ_0 , the approximation for φ_0 is

$$\varphi_0 \simeq (\sigma\Lambda/8\pi J) \ln(J/\lambda) \ll 1 \quad (14)$$

thus ensuring the validity of the linear approximation (8) in this case. Moreover, we have $|a\partial\varphi(r)/\partial r|_{r=a} \ll |\varphi_0|$ and thus the boundary conditions (13) are reasonable.

For small impurity concentrations c less than the characteristic value $c_0 = (a/\xi_0)^2$, magnetic clusters (9) of radius ξ_0 , produced by different impurity centres at mean separations $\bar{r} \simeq ac^{-1/2}$ in the plane, do not overlap with each other, and the ground-state configuration can be seen as a simple superposition of these clusters. The activation energy is still given by (10) for the regions outside the impurity clusters, constituting approximately $(1 - c/c_0)$ of the whole crystal volume. But thermic excitations within such clusters (occupying approximately c/c_0 of the whole volume) require less energy because the magnetization is turned, in these cases, against the asymptotic value $\varphi = 0$, on average, by $(\pi - \bar{\varphi})$ where

$$\bar{\varphi} \simeq \xi^{-2} \int d\mathbf{r} \varphi(r) \simeq \Lambda/2J$$

is the mean φ value in a single cluster. Thus we get an estimate for the Néel temperature as a function of c for low impurity concentrations $c \ll c_0$:

$$T_N(c)/T_N \simeq 1 - 2c\bar{\varphi}/\pi c_0 \simeq 1 - c/(\pi\sqrt{c_0 c_1}) \quad (15)$$

where $c_1 = J\lambda/\Lambda^2 \gg c_0$ is another characteristic concentration for the system. Note that the analogous formula for $\text{La}_2\text{CuO}_{4+\delta}$, obtained in [5], contains the linear concentrational dependence as c/c_1 .

It is seen from the estimates of (5) that c_0 in the real system is quite small: $c_0 \simeq 10^{-3}$, and in what follows we focus on the broad interval $c_0 \ll c \ll 1$ of concentrations where a great number of impurities occur, in the mean, in the fluctuation volume ξ^2 . Since the latter contribute to the spin anisotropy, which is effectively averaged over ξ^2 , the fluctuation length itself becomes a random function $\xi(\mathbf{r})$, depending on the random number of impurities of each type inside the volume $\xi^2(\mathbf{r})$ around \mathbf{r} . We can define $\xi(\mathbf{r})$ in a self-consistent manner as

$$\xi(\mathbf{r}) = a\sqrt{J/\lambda_\xi(\mathbf{r})} \quad (16)$$

where $\lambda_\xi(\mathbf{r})$ is the effective anisotropy constant, determined from the expansion of effective anisotropy energy

$$\begin{aligned} \mathcal{E}_{\text{eff}}(\mathbf{r}) = & -2S^2\{\lambda a^{-2} \cos^2 \varphi(\mathbf{r}) + \Lambda \xi^{-2}(\mathbf{r})[n_+ \cos^2(\varphi(\mathbf{r}) + \pi/4) \\ & + n_- \cos^2(\varphi(\mathbf{r}) - \pi/4)]\} \end{aligned} \quad (17)$$

near its minimum value $\mathcal{E}_{\text{min}}(\mathbf{r})$ so $\mathcal{E}_{\text{eff}}(\mathbf{r}) \simeq \mathcal{E}_{\text{min}}(\mathbf{r}) + \lambda_\xi(\mathbf{r})[\varphi(\mathbf{r}) - \varphi_0(\mathbf{r})]^2$ where

$$\tan 2\varphi_0(\mathbf{r}) = \Lambda(n_+ - n_-)a^2/\lambda\xi^2(\mathbf{r})$$

and n_\pm are the random numbers of impurities with $\sigma = \pm 1$ occurring inside $\xi(\mathbf{r})$. Hence we readily obtain

$$\lambda_\xi(\mathbf{r}) = \sqrt{\lambda^2 + \Lambda^2(n_+ - n_-)^2 a^4 \xi^{-4}(\mathbf{r})}. \quad (18)$$

For $c_0 \ll c \ll 1$ we can consider n_\pm as independent Gaussian random variables centred around $\bar{n}(\mathbf{r}) = c(\xi(\mathbf{r})/a)^2/2$ and possessing standard variance $[\bar{n}(\mathbf{r})]^{1/2}$. It is possible, in principle, to calculate the probability distribution function $P(c, \xi)$ for the realization of certain ξ at an arbitrary point \mathbf{r} at given c and then, integrating over all possible ξ , beginning from $\xi_{\text{min}} = \xi_0[1 + (\Lambda/\lambda)^2]^{-1/4} \ll \xi_0$ up to ξ_0 , consequently to obtain the probability for thermally activated turnover of arbitrary spin in a crystal with impurities. From obvious statistical reasoning we can simplify this consideration by taking into account only the most probable impurity configurations and by replacing the random quantity $(n_+ - n_-)^2$ with its mean value, that is $2\bar{n}(\mathbf{r})$, in (18). Then λ_ξ becomes independent of \mathbf{r} and from (16) we obtain an equation for the most probable fluctuation length ξ_c at given c :

$$(\xi_c/\xi_0)^4 + (c/c_1)(\xi_c/\xi_0)^2 - 1 = 0 \quad (19)$$

of which the solution

$$\xi_c = \xi_0 \left\{ [1 + (c/2c_1)^2]^{1/2} - c/2c_1 \right\}^{1/2} \quad (20)$$

is depicted in figure 2. Its asymptotic behaviour for $c \gg c_1$: $\xi_c \simeq \xi_0 \sqrt{c_1/c} \simeq \bar{r}J/\Lambda$ is similar to that found experimentally in $La_2CuO_{4+\delta}$ [15]. Accordingly we obtain for the most probable angle φ_c in the ground state:

$$\tan 2\varphi_c = [2c/((c^2 + 4c_1^2)^{1/2} - c)]^{1/2}$$

and for the most probable effective anisotropy: $\lambda_c = \lambda/\cos 2\varphi_c$.

In this approximation the system ground state can be seen as consisting fully of clusters of size ξ_c with magnetization directed with equal probability along $\pm\varphi_c$ at the centre of each cluster. Note that for all impurity concentrations under consideration these directions are contained within the sector $(\pi/4, -\pi/4)$ around $\varphi = 0$ and thus the ground state preserves the initial long-range order parameter, unlike in $La_2CuO_{4+\delta}$ where possible directions of magnetization in the ground state spread over the whole 2π range and long-range order vanishes at $c > c_1$ [5].

The minimum activation energy is provided by the energy difference between the ground-state configuration for the two adjacent clusters with $\varphi_1 = \varphi_c$, $\varphi_2 = -\varphi_c$ and their excited configuration, for instance, with $\varphi_1 = \varphi_c$, $\varphi_2 = -\varphi_c + \pi$ and amounts

$$E_{ac} \simeq J[(2\varphi_c - \pi)^2 - 4\varphi_c^2] / \ln(J/\lambda_c) \\ = E_{ac}^{(0)} [1 - (4/\pi)\varphi_c] / [1 + \ln \cos 2\varphi_c / \ln(J/\lambda)]. \quad (21)$$

Comparing (21) with (10), we finally arrive at the concentrational dependence of the Néel temperature at $c_0 \gg c \gg 1$:

$$T_N(c)/T_N \simeq \frac{1 - (2/\pi) \tan^{-1}[(\sqrt{1 + 4(c_1/c)^2} - 1)/2]^{-1/2}}{1 - \ln\{1 + 2/[\sqrt{1 + 4(c_1/c)^2} - 1]\} / [2 \ln(J/\lambda)]} \quad (22)$$

where the denominator differs from unity insignificantly over the whole concentration region. The formulae (15) and (22) agree well at $c \simeq c_0$ and thus provide a complete description for $T_N(c)$, that is for the system magnetic phase diagram, at all $c \ll 1$. The comparison of corresponding theoretical and experimental dependences (in normalized coordinates) in figure 3 demonstrates excellent agreement between them, and from the best fit for characteristic concentration $c_1 \simeq 6 \times 10^{-3}$ we can evaluate the effective number of Ni^{2+} spins affected by a single impurity centre as $n_{\text{eff}} \simeq 20$. This estimate qualitatively agrees with the value $\delta \simeq 0.13$ ($c \simeq 6.5\%$) at which the structural phase transition into the $Fm\bar{m}m$ phase is reported in $\text{La}_2\text{NiO}_{4+\delta}$ [7].

Relatively high values of $T_N(c)$ of order of 70–50 K at $c \geq c_1$ [10] can serve as a possible reason for the absence of a superconducting transition in this system, in contrast to La_2CuO_4 . This may be because the energy gain of approximately T_c on formation of a Cooper pair of charge carriers through the magnon-mediated interaction turns out to be less than the energy expenditure of approximately $T_N(c)$ required to break down the long-range magnetic order.

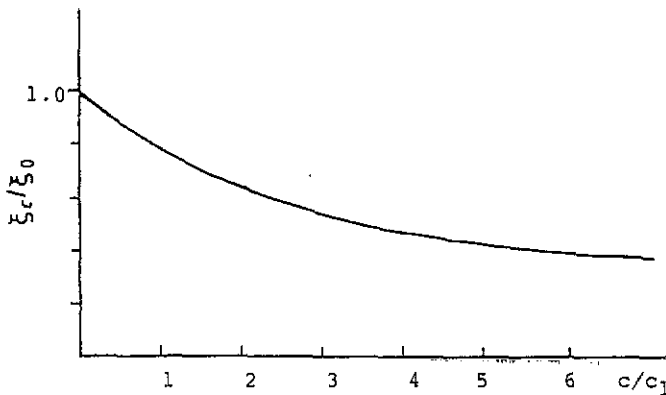


Figure 2. Characteristic length for magnetic fluctuations as a function of impurity concentration.

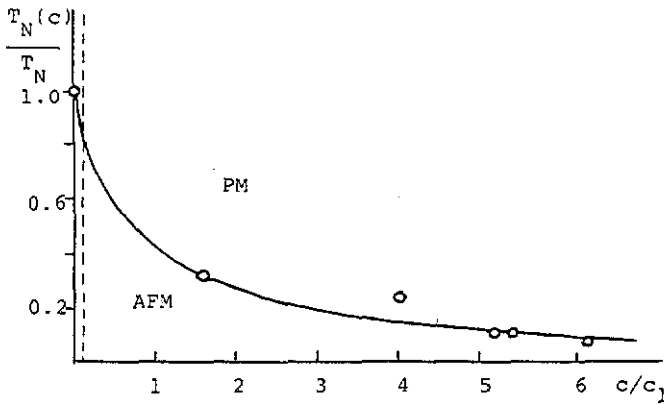


Figure 3. Magnetic phase diagram for the system $\text{La}_2\text{NiO}_{4+\delta}$ in normalized coordinates. The dots are taken from [10], the value $c_1 \simeq 6 \times 10^{-3}$ is chosen to provide the best fit to the experimental dependence. The dashed line represents $c_0/c_1 \ll 1$.

4. Conclusions

Based on the microscopic structure of spin interactions in the layered metal-oxide system $\text{La}_2\text{NiO}_{4+\delta}$, the continual model is developed to account for the influence of static deformations from superstoichiometric 'impurity' oxygens on in-plane spin anisotropy and thus on the stability of the magnetic long-range order in this system. At small impurity concentrations, when the mean distance \bar{r} between them exceeds the magnetic fluctuation length ξ , a linear $T_N(c)$ dependence is obtained. At $\bar{r} \ll \xi$ the fluctuation length and ground-state magnetic configurations as functions of c are found in a self-consistent approximation, taking into account the most probable statistical values of the impurity perturbation within the fluctuation length. The analytic formulae obtained for $T_N(c)$ give its consequent description throughout the concentration region $c \ll 1$ and agree well with available experimental data.

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