Nonlinear effect of perpendicular magnetic field on the antiferromagnetic phase transition in weakly coupled layered systems: Equal access decoupling scheme

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A system of equations of motion for the Green functions in layered antiferromagnets such as La_2CuO_4 and $YBa_2Cu_3O_6$ has been treated by an "equal access" decoupling scheme. The correlation functions are fully equivalent in this scheme in contrast with the ordinary random-phase approximation (RPA). The method provides a new insight into the nature of the RPA treatment of the localized spin dynamics in magnets. Explicit self-consistent expression for the sublattice magnetization in a perpendicular field is given. The dependence of the sublattice magnetization on a perpendicular magnetic field is studied. High temperature tails of the in-plane sublattice magnetization have been found to result from a nonlinear coupling of the perpendicular magnetic field with the antiferromagnetic order parameter.

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I. INTRODUCTION

Magnetic multilayered materials have received increasing attention in recent years as potential elements of magnetic devices and as a model system for fundamental theoretical studies. The high-temperature superconductors fall into this category of systems, the magnetic moments being practically confined within the copper-oxygen planes. The insulating phase of these systems provides much information about their magnetic dynamics. They are antiferromagnetic in normal state. It has already been established experimentally that they have a large difference in the antiferromagnetic coupling constant within CuO_2 planes and between the CuO_2 planes, i.e., in the *c* direction.[1] This remarkable anisotropy in the antiferromagnetic correlation is known to be at the origin of most of the unusual magnetic and electronic properties (see, e.g., Refs. 2 and 3 and references therein).

The Green function (GF) techniques allow one to relate observable quantities of a magnetic system with microscopic interaction parameters in the whole temperature range. The GF method was introduced into magnetic systems in 1959 by the pioneering work of Bogolyubov and Tyablikov who studied the thermodynamic properties of the spin-1/2 ferromagnetic systems.[4] Tahir-Kheli and ter Haar[5] extended successfully this technique to an arbitrary spin system in 1962. Since then, many authors have used this approach to study various kinds of magnetic systems.[6,7] The most remarkable advantage of the GF method is its approximate validity within the entire temperature range, which is not the case in the other approaches such as spin-wave theory, molecular-field theory, and high-temperature expansion theory. In this work we use a generalized form of the usual random-phase-approximation (RPA) procedure for layered magnets, which allows us to evaluate the sublattice magnetization and other characteristics.

We consider a three-dimensional (3D) anisotropic Heisenberg Hamiltonian on a two-sublattice system and use the double-time Green function technique to calculate the magnetization on each sublattice as well as the Néel temperature. The present work differs from previous calculations[2,3] in that we have employed the equal accesses (EA) decoupling scheme for linearizing higher-order Green functions.[8] The magnetic field included in the Hamiltonian was applied perpendicular to the CuO_2 planes and at the same time perpendicular to the direction of the spontaneous magnetization of the sublattices. Our motivation here is to see the effect of the thermal average of the transverse component of the spin operator on the sublattice magnetization as well as its effect on the Néel temperature.

The plan of our paper is as follows. In Sec. II we present the Hamiltonian and derive a set of coupled equations for the Green function using the equal accesses decoupling scheme. A closed expressions for the sublattice magnetization and for the Néel temperature are also reported. In Sec. III we present numerical results of our calculations and compare them with those obtained with the use of RPA.[9] Finally, in Sec. IV, we discuss the validity of the new decoupling scheme and the physical consequences of the nonlinear coupling of the external magnetic field with the sublattice magnetization.

II. THEORETICAL MODEL

We divide the simple cubic lattice into two sublattices, so that all the nearest neighbors of every site belonging to one sublattice belong to the other sublattice. The Hamiltonian of the present problem then is that of a 3D anisotropic Heisenberg model and reads:

$$\mathcal{H} = \sum_{ab} 2J_{\parallel}\mathbf{S}_l \cdot \mathbf{S}_m + \sum_{c} 2J_{\perp}\mathbf{S}_l \cdot \mathbf{S}_m - \gamma H \sum_{f} S_l^x - \gamma H \sum_{g} S_m^x, \tag{1}$$

where *ab* and *c* denote the nearest-neighbor summation within and perpendicular to the CuO_2 planes, in the *c* direction, respectively. Each plane perpendicular to the c direction contains an equal number of sites belonging to both lattices. Thus every separate plain forms a [two-dimensional (2D)] antiferromagnetic system. The parameters $J_{\parallel}$ and $J_{\perp}$ are the antiferromagnetic coupling constants in the ab plane and in the c direction, respectively. On assuming the two-sublattice model and nearest-neighbor interaction, f and g denote the sites of the two sublattices with different spin orientations [spin up (f) and down (g)]. The nearest neighbor of a lattice site belonging to sublattice f belongs to the g sublattice and vice versa. The spontaneous magnetization of each layer lies within the plane of the layer and thus defines the z direction. The magnetic field H is applied perpendicular to the planes and its direction points out the x direction. The quantity $\gamma = g\mu_B$ where μ_B is the Bohr magneton and g is the usual g factor. To investigate the Hamiltonian given in Eq. (1) the following (retarded) Green's functions have been introduced (in standard notation for spin operators):

$$G^{+-}_{lf,mf}(t,t') \equiv \langle\langle S^{+}_{lf}(t) | S^{-}_{mf}(t')\rangle\rangle = -i\theta(t-t')\langle [S^{+}_{lf}(t), S^{-}_{mf}(t')]\rangle_T, \tag{2a}$$

similarly:

$$G^{+-}_{lg,mf}(t,t') \equiv \langle\langle S^{+}_{lg}(t) | S^{-}_{mf}(t')\rangle\rangle, \tag{2b}$$

$$G^{--}_{lf,mf}(t,t') \equiv \langle\langle S^{-}_{lf}(t) | S^{-}_{mf}(t')\rangle\rangle, \tag{2c}$$

$$G^{--}_{lg,mf}(t,t') \equiv \langle\langle S^{-}_{lg}(t) | S^{-}_{mf}(t')\rangle\rangle, \tag{2d}$$

$$G^{z-}_{lf,mf}(t,t') \equiv \langle\langle S^{z}_{lf}(t) | S^{-}_{mf}(t')\rangle\rangle, \tag{2e}$$

$$G^{z-}_{lg,mf}(t,t') \equiv \langle\langle S^{z}_{lg}(t) | S^{-}_{mf}(t')\rangle\rangle, \tag{2f}$$

where $S^{\pm} = S^{x} \pm iS^{y}$ are the spin rising and spin lowering operators, respectively, with usual commutation relations: $[S^{+}_{l}, S^{-}_{m}] = 2S^{z}_{l}\delta_{lm}$ and $[S^{z}_{l}, S^{\pm}_{m}] = \pm S^{\pm}_{l}\delta_{lm}$, and a suffix (e.g.) lf means a site l in the sublattice f. The equations of motion for the above Green's functions, Fourier transformed with respect to time, have been derived in the usual way,[8]

$$\begin{aligned}\omega G^{+-}_{lf,mf} = {} & \langle [S^{+}_{lf}, S^{-}_{mf}]\rangle \frac{\delta_{lm}}{2\pi} + 2J_{\parallel}\sum_{\delta}^{ab} \langle\langle S^{z}_{lf}S^{+}_{l+\delta,g} | S^{-}_{mf}\rangle\rangle \\ & - \langle\langle S^{+}_{lf}S^{z}_{l+\delta,g} | S^{-}_{mf}\rangle\rangle + 2J_{\perp}\sum_{\delta'}^{c} \langle\langle S^{z}_{lf}S^{+}_{l+\delta',g} | S^{-}_{mf}\rangle\rangle \\ & - \langle\langle S^{+}_{lf}S^{z}_{l+\delta',g} | S^{-}_{mf}\rangle\rangle - \gamma H G^{z-}_{lf,mf},\end{aligned} \tag{3a}$$

$$\begin{aligned}\omega G^{+-}_{lg,mf} = {} & 2J_{\parallel}\sum_{\delta}^{ab} \langle\langle S^{z}_{lg}S^{+}_{l+\delta,f} | S^{-}_{mf}\rangle\rangle - \langle\langle S^{+}_{lg}S^{z}_{l+\delta,f} | S^{-}_{mf}\rangle\rangle \\ & + 2J_{\perp}\sum_{\delta'}^{c} \langle\langle S^{z}_{lg}S^{+}_{l+\delta',f} | S^{-}_{mf}\rangle\rangle \\ & - \langle\langle S^{+}_{lg}S^{z}_{l+\delta',f} | S^{-}_{mf}\rangle\rangle - \gamma H G^{z-}_{lg,mf},\end{aligned} \tag{3b}$$

$$\begin{aligned}\omega G^{z-}_{lf,mf} = {} & \langle [S^{z}_{lf}, S^{-}_{mf}]\rangle \frac{\delta_{lm}}{2\pi} + 2J_{\parallel}\sum_{\delta}^{ab} \langle\langle S^{+}_{lf}S^{-}_{l+\delta,g} | S^{-}_{mf}\rangle\rangle \\ & - \langle\langle S^{-}_{lf}S^{+}_{l+\delta,g} | S^{-}_{mf}\rangle\rangle + J_{\perp}\sum_{\delta'}^{c} \langle\langle S^{+}_{lf}S^{-}_{l+\delta',g} | S^{-}_{mf}\rangle\rangle \\ & - \langle\langle S^{-}_{lf}S^{+}_{l+\delta',g} | S^{-}_{mf}\rangle\rangle - (\gamma H/2)(G^{+-}_{lf,mf} - G^{--}_{lf,mf}),\end{aligned} \tag{3c}$$

$$\begin{aligned}\omega G^{z-}_{lg,mf} = {} & 2J_{\parallel}\sum_{\delta}^{ab} \langle\langle S^{+}_{lg}S^{-}_{l+\delta,f} | S^{-}_{mf}\rangle\rangle - \langle\langle S^{-}_{lg}S^{+}_{l+\delta,f} | S^{-}_{mf}\rangle\rangle \\ & + J_{\perp}\sum_{\delta'}^{c} \langle\langle S^{+}_{lg}S^{-}_{l+\delta',f} | S^{-}_{mf}\rangle\rangle - \langle\langle S^{-}_{lg}S^{+}_{l+\delta',f} | S^{-}_{mf}\rangle\rangle \\ & - (\gamma H/2)(G^{+-}_{lg,mf} - G^{--}_{lg,mf}),\end{aligned} \tag{3d}$$

$$\begin{aligned}\omega G^{--}_{lf,mf} = {} & 2J_{\parallel}\sum_{\delta}^{ab} \langle\langle S^{-}_{lf}S^{z}_{l+\delta,g} | S^{-}_{mf}\rangle\rangle - \langle\langle S^{z}_{lf}S^{-}_{l+\delta,g} | S^{-}_{mf}\rangle\rangle \\ & + 2J_{\perp}\sum_{\delta'}^{c} \langle\langle S^{-}_{lf}S^{z}_{l+\delta',g} | S^{-}_{mf}\rangle\rangle \\ & - \langle\langle S^{z}_{lf}S^{-}_{l+\delta',g} | S^{-}_{mf}\rangle\rangle + (\gamma H) G^{z-}_{lf,mf},\end{aligned} \tag{3e}$$

$$\begin{aligned}\omega G^{--}_{lg,mf} = {} & 2J_{\parallel}\sum_{\delta}^{ab} \langle\langle S^{-}_{lg}S^{z}_{l+\delta,f} | S^{-}_{mf}\rangle\rangle - \langle\langle S^{z}_{lg}S^{-}_{l+\delta,f} | S^{-}_{mf}\rangle\rangle \\ & + 2J_{\perp}\sum_{\delta'}^{c} \langle\langle S^{-}_{lg}S^{z}_{l+\delta',f} | S^{-}_{mf}\rangle\rangle \\ & - \langle\langle S^{z}_{lg}S^{-}_{l+\delta',f} | S^{-}_{mf}\rangle\rangle + (\gamma H) G^{z-}_{lg,mf}.\end{aligned} \tag{3f}$$

Until now the calculations are entirely exact, but the equations of motion (3) contain a three-point GF's such as $\langle\langle S^{\alpha}_{l}S^{\beta}_{m}, S^{-}_{m'}\rangle\rangle, \alpha,\beta = +,-,z$, which are difficult to determine. A direct generalization of the original RPA decoupling[8] then allows one to write:

$$\langle\langle S^{\alpha}_{l}S^{\beta}_{m}, S^{-}_{m'}\rangle\rangle = \langle S^{\alpha}_{l}\rangle_T G^{\beta-}_{m,m'} + \langle S^{\beta}_{m}\rangle_T G^{\alpha-}_{l,m'}, \quad \alpha,\beta = +,-,z, \tag{4}$$

where $l \neq m, \langle x\rangle_T = \mathrm{tr}[x\exp(-\beta\mathcal{H})]/\mathrm{tr}[\exp(-\beta\mathcal{H})]$, is the thermal average of the operator x, $\mathcal{H}$ is the Hamiltonian, and T is the temperature of the system. The merit in this generalized form of decoupling is that both correlation functions: $\langle S^{\beta}_{l}, S^{-}_{m}\rangle, \langle S^{\alpha}_{l}, S^{-}_{m}\rangle$ enter the equation of motion in a completely symmetrical way, or in other words, have equal access. That is why we can call it the "equal access" (EA) decoupling (EA-RPA). For the system of spins coupled identically—the Kittel-Shore-Kac model magnet[10,11]—the magnetization and the magnetic susceptibility calculated within the present EA-RPA approach do agree with the results of exact calculations.[12,13] It provides an interesting insight into the nature of the EA-RPA treatment of the localized spin dynamics in magnets. It is also worth mentioning that the magnon conductivity, which depends upon the decoupling scheme, is modified appreciably by the different decoupling parameters and by the anisotropy of the system.[14]

Employing the above decoupling approach and introducing the space Fourier transformation of the resulting Green functions, we arrive at the nonuniform system of linear algebraic equations for the Green functions we seek

$$(\omega - \eta\overline{S})G_{1k}^{+-} - \zeta\overline{S}G_{2k}^{+-} - h_g^{+}G_{1k}^{z-} + \zeta\langle S_{lf}^{+}\rangle_T G_{2k}^{z-} + (\gamma H)G_{1k}^{z-} = \overline{S}/\pi, \tag{5a}$$

$$(\omega + \eta\overline{S})G_{2k}^{+-} + \zeta\overline{S}G_{1k}^{+-} - h_f^{+}G_{2k}^{z-} + \zeta\langle S_{lg}^{+}\rangle_T G_{1k}^{z-} + (\gamma H)G_{2k}^{z-} = 0, \tag{5b}$$

$$\omega G_{1k}^{z-} - (1/2)\zeta\langle S_{lf}^{+}\rangle_T G_{2k}^{--} - (1/2)h_g^{-}G_{1k}^{+-} + (1/2)h_g^{+}G_{1k}^{--} + (1/2)\zeta\langle S_{lf}^{-}\rangle_T G_{2k}^{+-} + (\gamma H/2)(G_{1k}^{+-} - G_{1k}^{--}) = -\langle S_{lf}^{-}\rangle_T/2\pi, \tag{5c}$$

$$\omega G_{2k}^{z-} - (1/2)\zeta\langle S_{lg}^{+}\rangle_T G_{1k}^{--} - (1/2)h_f^{-}G_{2k}^{+-} + (1/2)h_f^{+}G_{2k}^{--} + (1/2)\zeta\langle S_{lg}^{-}\rangle_T G_{1k}^{+-} + (\gamma H/2)(G_{2k}^{+-} - G_{2k}^{--}) = 0, \tag{5d}$$

$$(\omega + \eta\overline{S})G_{1k}^{--} - \zeta\langle S_{lf}^{-}\rangle_T G_{2k}^{z-} + \zeta\overline{S}G_{2k}^{--} + h_g^{-}G_{1k}^{z-} - (\gamma H)G_{1k}^{z-} = 0, \tag{5e}$$

$$(\omega - \eta\overline{S})G_{2k}^{--} - \zeta\langle S_{lg}^{-}\rangle_T G_{1k}^{z-} - \zeta\overline{S}G_{1k}^{--} + h_f^{-}G_{2k}^{z-} - (\gamma H)G_{2k}^{z-} = 0, \tag{5f}$$

where $\eta = 8J_\parallel + 4J_\perp$, $\zeta = 2J_\parallel\Sigma_\delta\, e^{ik\cdot\delta} + 2J_\perp\Sigma_{\delta'}\, e^{ik\cdot\delta'}$, and $\overline{S} = \langle S_{lf}^{z}\rangle_T = -\langle S_{lg}^{z}\rangle_T$ (the sublattice magnetization is related to $\overline{S}$ by $M = \gamma\overline{S}$),

$$h_{f(g)}^{\pm} = 2J_\parallel\sum_\delta \langle S_{l+\delta,f(g)}^{\pm}\rangle_T + 2J_\perp\sum_{\delta'}\langle S_{l+\delta',f(g)}^{\pm}\rangle_T = \eta\langle S_{f(g)}^{\pm}\rangle_T,$$

($h_{f(g)}^{+(-)}$ is the local field parameter, see Ref. 13),

$$G_{lf,mf}^{+-} = (2/N)\sum_k G_{1k}^{+-}e^{ik\cdot(l-m)}, \tag{6}$$

$$G_{lg,mf}^{+-} = (2/N)\sum_k G_{2k}^{+-}e^{ik\cdot(l-m)}, \tag{7}$$

where N is the total number of spins in the lattice and k is the reciprocal lattice vector which runs over the first Brillouin zone. G_{1k} is the Green's function when l and m are on the same sublattice and G_{2k} when they are on different sublattices.

Solving the system of equations (5) one finds the following form for G_{1k}^{+-}:

$$G_{1k}^{+-} = \frac{A\omega^2 + B\omega + C}{2\pi\omega(\omega^2 - D^2)}, \tag{8}$$

where

$$A = 2\overline{S},$$

$$B = \eta(\langle S_{lf}^{x}\rangle_T^2 + \langle S_{lf}^{y}\rangle_T^2 + \langle S_{lf}^{z}\rangle_T^2) + \langle S_{lf}^{z}\rangle_T^2 + \langle S_{lf}^{-}\rangle_T\gamma H,$$

$$C = -\eta\overline{S}\langle S_{lf}^{+}\rangle_T\gamma H,$$

and

$$D^2 = (\eta^2 - \zeta^2)(\langle S_{lf}^{x}\rangle_T^2 + \langle S_{lf}^{y}\rangle_T^2 + \langle S_{lf}^{z}\rangle_T^2) + 2\eta\langle S_{lf}^{x}\rangle_T\gamma H. \tag{9}$$

Using the spectral intensity theorem of the Green function theory[4] we have calculated the correlation function $\langle S_{lf}^{-}S_{lf}^{+}\rangle$ as

$$\langle S_{lf}^{-}S_{lf}^{+}\rangle = \frac{2}{N}\sum_k\left[\frac{\eta(\langle S_{lf}^{x}\rangle_T^2 + \langle S_{lf}^{y}\rangle_T^2 + 2\langle S_{lf}^{z}\rangle_T^2) + \langle S_{lf}^{-}\rangle_T\gamma H}{2D}\times\coth\left(\frac{D}{2k_BT}\right) - \overline{S}\right]. \tag{10}$$

For spin-1/2 we have $S_l^z = 1/2 - S_l^- S_l^+$, therefore $\overline{S}$ can be expressed in terms of the correlation function $\langle S_{lf}^{-}S_{lf}^{+}\rangle$ (after dropping the suffix lf, T) as

$$\overline{S} = \left[2 + \frac{4}{N}\sum_k\left(\frac{\eta\left[2 + \left(\frac{\langle S^x\rangle}{\langle S^z\rangle}\right)^2 + \left(\frac{\langle S^y\rangle}{\langle S^z\rangle}\right)^2\right] + \frac{\langle S^-\rangle\gamma H}{\langle S^z\rangle^2}}{2\sqrt{(\eta^2 - \zeta^2)\left[1 + \left(\frac{\langle S^x\rangle}{\langle S^z\rangle}\right)^2 + \left(\frac{\langle S^y\rangle}{\langle S^z\rangle}\right)^2\right] + 2\eta\frac{\langle S^x\rangle\gamma H}{\langle S^z\rangle^2}}}\coth\left(\frac{D}{2k_BT}\right) - 1\right)\right]^{-1}. \tag{11}$$

This is a self-consistent equation for $\overline{S}$: it gives us the variation of the sublattice magnetization with temperature. It can be reduced to the expression for the magnetization given in Ref. 9 by putting $H = 0$ and $\langle S^x\rangle = \langle S^y\rangle = 0$. Other types of self-consistent solutions can also be extracted from Eq. (11), e.g., $\langle S^x\rangle = \langle S^y\rangle = \langle S^z\rangle = 0$, which corresponds to the paramagnetic state. These states may be unstable (metastable) in some range of temperature. The Néel temperature T_N can be extracted from Eq. (11) using the well-known property $\overline{S} \to 0$ as $T \to T_N$, at $H=0$, a simple algebra leads to the following expression for the Néel temperature:

$$T_N = \frac{1}{4k_B}\left[\frac{1}{N}\sum_k \left(1+\frac{\overline{S}^2}{h^2}\right)\left(\frac{\eta}{\eta^2-\zeta^2}\right)\right]^{-1}, \tag{12}$$

where

$$h = \sqrt{\langle S^x\rangle^2 + \langle S^y\rangle^2 + \langle S^z\rangle^2} \tag{13}$$

is the local field parameter.[13] One might think of this parameter as a sort of scaling factor depending on temperature in a complicated way via the thermal averages. The above formula for the Néel temperature can be reduced (in our geometry where the component $\langle S^y\rangle$ vanishes) to the expression:

$$T_N = \frac{J_\parallel}{4k_B\left(1+\dfrac{1}{1+(\langle S^x\rangle/\langle S^z\rangle)^2}\right)I(r)}, \tag{14}$$

where $r=J_\perp/J_\parallel$ and

$$I(r) = \frac{2}{N}\sum_k \frac{(2+r)}{(2+r)^2-[\cos(k_x a)+\cos(k_y a)+r\cos(k_z c)]^2}. \tag{15}$$

We apply here the well-known linear relation between $\langle S^x\rangle$ and H: $\langle S^x\rangle=\chi_{11}H$, where χ_{11} stands for the xx component of the magnetic susceptibility tensor. In this geometry the component $\langle S^y\rangle$ vanishes. The value of χ_{11} has been put $\chi_{11}=\gamma/[8J_\parallel(2+r)]$ as it follows from Ref. 15. One should remember that contrary to the parallel component χ_{33}, the x component χ_{11} does not show a significant temperature dependence. As it can be seen from Eqs. (9) and (11) the perpendicular magnetic field H enters the self-consistent equation in the second power only so that its effect on the magnetization $\langle S^z\rangle$ is intrinsically nonlinear. A mathematical manipulation leads to the following form for the Néel temperature (see the Appendix):

$$T_N = \frac{J_\parallel}{4k_B\left(1+\dfrac{1}{1+(\gamma H/[8J_\parallel\overline{S}(2+r)])^2}\right)[0.5055-0.162\ln(r)]}. \tag{16}$$

Let us mention that a similar logarithmic term has been previously obtained by Singh *et al.*[2] The above formula for T_N differs from Eq. (12) of Ref. 9 in that a nonlinear field dependent term appears (due to the EA-RPA decoupling scheme) in the denominator. The impact of such a term will be discussed in the next section. It is clear also from Eq. (16) that when $r\to 0$ the Néel temperature $T_N\to 0$. This is in agreement with the Mermin and Wagner theorem that there cannot be long-range order in two-dimensional systems at finite temperature.

III. NUMERICAL CALCULATION OF THE SUBLATTICE MAGNETIZATION FOR $YBa_2Cu_3O_{6+x}$ AND La_2CuO_4.

In what follows we use Eq. (11) to study the effect of the magnetic field applied perpendicular to the direction of the sublattice magnetization onto the spontaneous magnetization within each sublattice. The temperature dependence of the sublattice magnetization $\langle S^z\rangle$ at the presence of the perpendicular magnetic field H has been obtained numerically with the summation in Eq. (11) replaced by a triple integral over the Brillouin zone. The integral was evaluated with a step adaptive iterative method. Then the value of the magnetization was set up and the corresponding temperature was found by iterations until a convergence better then 0.1% was attained.

Figure 1(a) presents the variation of the sublattice magnetization $M(T)$, normalized to its value at zero temperature $M(0)$, as a function of temperature at $H=0$. The plot is given for the undoped $YBa_2Cu_3O_{6+x}$ (solid curve) and La_2CuO_4 (dashed curve). The value of $J_\parallel$ is put to 0.006 and 0.009 eV, respectively, which at the ratio $r=J_\perp/J_\parallel$ equal to 10^{-5} (see Ref. 9) reproduces experimental Néel temperatures for both compounds. One should notice a slight difference between the values of $J_\parallel$ found here and those reported in literature. Let us mention, however, that some authors consider $2J_\parallel$ instead of $J_\parallel$ and $2J_\perp$ instead of $J_\perp$.[9] There is also a hesitation in the literature[9] about the value of the perpendicular/parallel coupling ratio $r=J_\perp/J_\parallel$. To illustrate the extent to which this quantity influences the sublattice magnetization we show in Fig. 1(b) the results obtained with $J_\parallel=0.006$ eV for three different values of r. The plot is given for the undoped $YBa_2Cu_3O_{6+x}$. One can see an upwards shift of the Néel temperature with increasing r. Let us mention that the interplaner coupling $J_\perp$, though very small, is essential for a system realizing long-range order at nonzero temperature.

The temperature dependence of the sublattice magnetization at the presence of the perpendicular magnetic field ($H\neq 0$) is represented in Figs. 2 and 3 for $YBa_2Cu_3O_{6+x}$ and

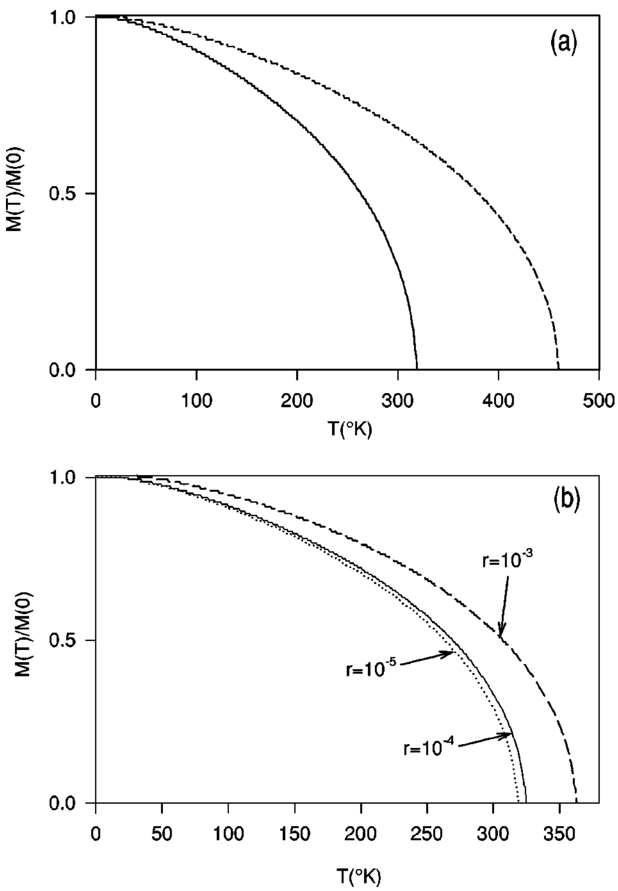

FIG. 1. (a) Variation of the sublattice magnetization $M(T)$ [normalized to its value at zero temperature $M(0)$] as a function of temperature for the undoped $YBa_2Cu_3O_{6+x}$ (solid curve) and La_2CuO_4 (dashed curve) at $H=0$. The value of $J_\parallel$ is put to 0.006 and 0.009 eV, respectively, and the ratio $r=10^{-5}$. (b) Variation of the sublattice magnetization $M(T)/M(0)$ vs temperature for the undoped $YBa_2Cu_3O_{6+x}$ and for $r=10^{-5}$ (doted curve), $r=10^{-4}$ (solid curve), and $r=10^{-3}$ (dashed curve).

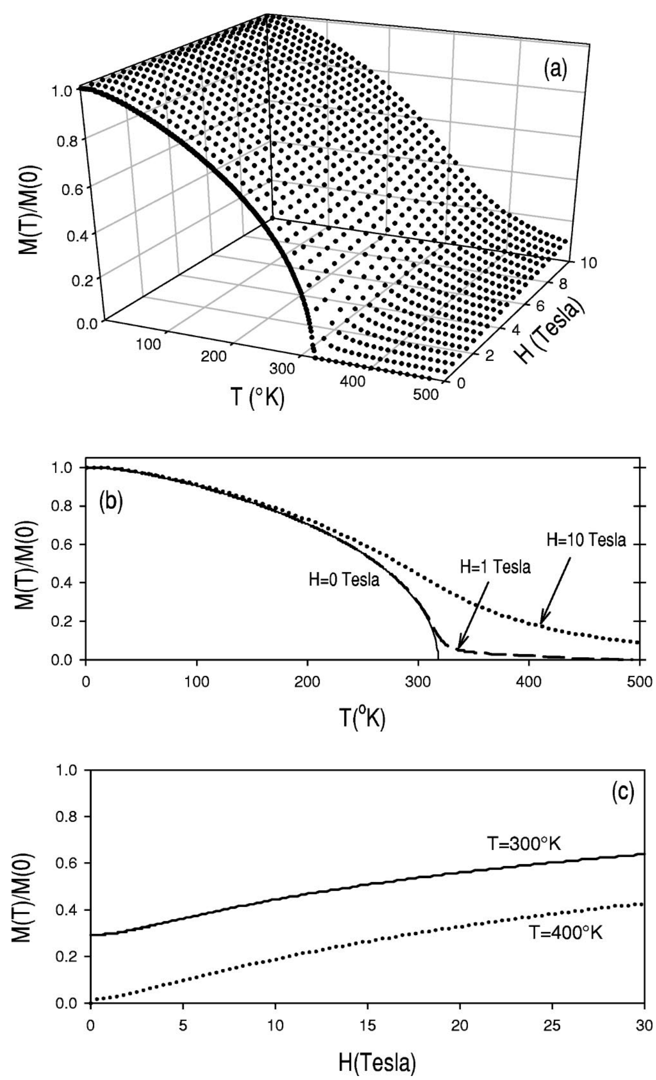

FIG. 2. (a) Three-dimensional plot of the sublattice magnetization $M(T)/M(0)$ as a function of temperature and of perpendicular field H for the undoped $YBa_2Cu_3O_{6+x}$. (b) Sublattice magnetization $M(T)/M(0)$ as a function of temperature for different values of the perpendicular field $H=0$ T (solid curve), $H=1$ T (dashed curve), and $H=10$ T (doted curve). Notice the high temperature tails of the sublattice magnetization above the Nèel temperature (for a nonzero value of the magnetic field). (c) Sublattice magnetization $M(T)/M(0)$ as a function of H for two different values of temperature. The solid curve for $T=300$ K (which is below the Néel temperature) and the doted curve for $T=400\ \mathrm{K} > T_N$. The nonzero value of the magnetization (at $H=0$) for the former and the zero value for the latter is noteworthy. The value of $J_\parallel=0.006$ eV, and the ratio $r=10^{-5}$ is kept the same for (a)–(c).

La_2CuO_4, respectively. The other parameters are the same as in Fig. 1(a). Generally, the perpendicular magnetic field smears out the phase transition in analogy to a nonzero field conjugated to the order parameter. A similar effect is often reported in many second order phase transitions in the absence of any symmetry breaking field and is explained by various phenomena such as the presence of impurities or logarithmic corrections to the critical behavior.[16,17] In the present case one can expect such smearing out of the phase transition even in the perfect $YBa_2Cu_3O_{6+x}$ and La_2CuO_4 crystals in the magnetic field with a nonzero component perpendicular to the CuO_2 planes. On the other hand the same phenomenon may be expected at the field oriented parallel to the sublattice magnetization, e.g., in the magnetic susceptibility measurements, if the CuO_2 planes are not strictly parallel to each other due to some structural defects.

IV. DISCUSSION AND CONCLUSION

The random-phase approximation (RPA) of Tyablikov,[4] is explicitly aimed at the description of ferromagnets and leads directly to the mean field results. It is the simplest and most current decoupling scheme, but it results in a disagreement with the low temperature theory,[18] where only long-wavelength spin waves are excited, and in turn, the local magnetization direction varies slowly through the crystal.[19] The equal-access random-phase approximation (EA-RPA) introduced in this paper is a natural extension of the RPA, conserving more information on the spin-spin correlation functions than the original RPA description. It covers a mul-

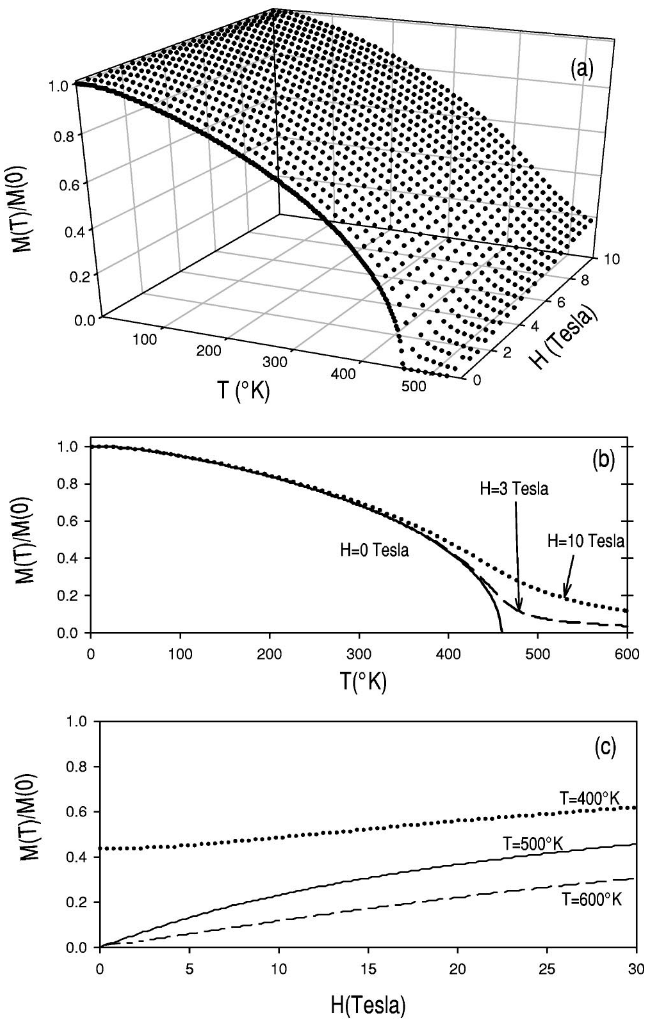

FIG. 3. (a) The same as in Fig. 2(a) but for La_2CuO_4. (b) The same as in Fig. 2(b) but for $H=0$ T (solid curve), $H=3$ T (dashed curve), and $H=10$ T (doted curve). (c) The same as in Fig. 2(c) but for $T=400$ K (doted curve), $T=500$ K (solid curve), and $T=600$ K (dashed curve). The value of $J_{\parallel}=0.009$ eV, and the ratio $r=10^{-5}$ is kept the same for (a)–(c).

titude of spin systems including antiferromagnets, but its basic physics and range is of the mean field approximation type. It can probably be improved to adjust its critical indices to the indices of the static scaling theory using the method developed by Czachor and Holas[20] for the RPA.

To study the magnetic properties in layered antiferromagnets one has to take into account a greater variety of spin-spin correlations than in the case of ferromagnets. To do so, we have first introduced a generalized RPA decoupling [Eq. (4)] of the three-spin correlation functions (Green's functions) involved. We claim that in doing so we have entered a reasonable way towards improving the GF method for the magnetization calculations in layered structures. A way more systematic than those quoted in the Introduction. On assuming then an equivalent and symmetric role of two different two-spin correlation functions pertinent to the problem one has the symmetric "equal-access" form of the RPA decoupling (EA-RPA). At this step one obtains the canonical set [Eq. (5)] of six equation of motion for the six GF's we seek. The system of equations [Eq. (5)] is solved analytically and an explicit expression [Eq. (8)] for the Green function has been found. On looking at the roots of its denominator one can find a formula for the dispersion relations of magnetic excitations of the system. Such a formula depends on the local field parameters $h_{f(g)}^{+(-)}$ which allow one to predict the dispersion relations for the antiferromagnetic excitations, such as they can be seen in neutron scattering. The spectral intensity theorem of the Green function theory[4] and Eq. (8) has been used to calculate the sublattice magnetization [Eq. (11)]. The expression is found to involve all the components of the magnetization so that the external magnetic field in the x direction couples in the second order to the z component of the magnetization which is the order parameter in the phase transition under study. The analytical expression for the Néel temperature Eq. (12) is then found in terms of the parameters of the model and, what is more interesting, on the local field parameter h.[13]

As an example of an application of the new EA-RPA scheme we have studied here the effect of the magnetic field perpendicular to the spontaneous magnetization in the CuO_2 layers onto the behavior of the antiferromagnetic order parameter. The magnetic field enters the equations in the second power and therefore this is a second order effect. The application of such a field results in high temperature tails of the sublattice magnetization above the Nèel temperature. Thus the effect is analogous to the linear effect of the field conjugated with the order parameter. Similar high temperature tails are often encountered in second order phase transitions of different nature. Their explanation invokes various phenomena ranging from impurities to so-called logarithmic corrections to the critical behavior.[16,17] Here the origin of the tails is the existence of the nonlinear coupling between the sublattice magnetization and the perpendicular macroscopic field.

The results obtained in the present work should be compared with those obtained with the use of renormalization group (RG) and $1/N$ expansion for a similar model.[21] The RG approach implies an expansion of the Hamiltonian as a function of deviation from the ordered state and, therefore, is well adapted to low temperatures where the deviations are indeed small. On the other hand the $1/N$ expansion (N the dimension of the spin variable, $N=3$ for the Heisenberg model) should in principle work in the whole range of temperatures. It turns out, however, that the first order expansion overestimates the sublattice magnetization at low temperatures if there are non-negligible anisotropic terms in the initial Hamiltonian (Fig. 2 of Ref. 21). Both approaches give a double logarithmic expression for the Nèel temperature [Eqs. (45) and (87) of Ref. 21]. A renormalization of the Nèel temperature is necessary when treating experimental data in both cases. A crossover temperature can be discerned between two regimes, although the magnetization curve rests continuous and smooth.

Compared with those approaches the present work gives a unique expression for the whole temperature range. The starting Hamiltonian does not contain anisotropy terms so that a direct comparison can be made for La_2CuO_4 only (Fig. 1 of Ref. 21), where the anisotropy parameters are weak.

The results of Ref. 22 suggest that a simple rescaling of the temperature may give correct results in analogy to those obtained in Ref. 21 with a proper renormalization of the Nèel temperature. Comparison of our Fig. 2(b) with Fig. 1 of Ref. 21 suggests that the general shape of the magnetization curve at $H=0$ is correct for weak anisotropy. Moreover, introduction of anisotropy terms is fairly analogous to the one-particle energy contribution of Ref. 22. Therefore it is expected that the EA-RPA for a Hamiltonian with nonzero anisotropy will produce a magnetization curve similar to Fig. 2 of Ref. 21 with a characteristic crossover from a rather weak temperature dependence at low temperatures to a steep increase directly below T_N. We now work on the formulation of the EA-RPA with anisotropy terms.

APPENDIX

Starting from Eq. (15) for $I(r)$. After using the method of partial fractions and changing the summation over k into integration, Eq. (15) reduces to

$$I(r) = \frac{V}{\pi^3}\int_0^{\pi/c}\int_0^{\pi/a}\int_0^{\pi/a} \times \frac{dk_x dk_y dk_z}{[2-\cos(k_x a)-\cos(k_y a)] + r[1-\cos(k_z c)]}, \quad \text{(A1)}$$

where V stands for the volume of the unit cell. Changing to the new variables α,β,γ via the transform $k_x a=\alpha, k_y a=\beta, k_z c=\gamma$, integrating first over γ and then over β makes the above equation changing to the form:

$$I(r) = \int_0^{\pi} A(\alpha,r)B(\alpha,r)du, \quad \text{(A2)}$$

where

$$A(\alpha,r) = 2[(3-\cos\,\alpha)(2r+1-\cos\,\alpha)]^{-1/2} \quad \text{(A3)}$$

and

$$B(\alpha,r) = \int_0^{\pi/2}(1-\Gamma\,\sin^2\theta)^{-1/2}d\theta, \quad \text{(A4)}$$

where

$$\Gamma = 4r[(3-\cos\,\alpha)(r+1-\cos\,\alpha)]^{-1}. \quad \text{(A5)}$$

The value of the integral in Eq. (A2) becomes $\pi/2$ at very small r and the leading contribution to the integral comes from small values of α. The integral in Eq. (A2) has been calculated numerically as a function of the ratio r. The results shows that its value can be approximately written as $I(r)=[0.5055-0.162\,\ln(r)]$. The Néel temperature Eq. (16) results directly from inserting the value of $I(r)$ into Eq. (14).

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[1] J. M. Tranquada, A. H. Moudden, A. I. Goldman, P. Zolliker, D. E. Cox, G. Shirane, S. K. Sinha, D. Vaknin, D. C. Johnston, M. S. Alvarez, A. J. Jacobson, J. T. Lewandowski, and J. M. Newsam, Phys. Rev. B **38**, 2477 (1988).

[2] A. Singh, Z. Tesanovic, H. Tang, G. Xiao, C. L. Chien, and J. C. Walker, Phys. Rev. Lett. **64**, 2571 (1990).

[3] A. Pratap, R. S. Ajay, and R. S. Tripathi, Phys. Status Solidi B **197**, 453 (1996).

[4] N. N. Bogolyubov and S. V. Tyablikov, Dokl. Akad. Nauk SSSR **126**, 53 (1959) [Sov. Phys. Dokl. **4**, 606 (1959)] ; S. V. Tyablikov, Ukr. Mat. Zh. **11**, 187 (1959). A review of Green's functions and of Tyablikov's application of them to ferromagnetism is given by D. N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960) [Sov. Phys. Usp. **3**, 320 (1960)].

[5] R. Tahir-Kheli and D. Ter Harr, Phys. Rev. **127**, 88 (1962).

[6] M. E. Lines, Phys. Rev. **156**, 534 (1967).

[7] H. B. Callen, Phys. Rev. **130**, 890 (1963); F. Burr Anderson and H. B. Callen, *ibid.* **136**, A1068 (1964); R. A. Tahir-Kheli, Phys. Rev. B **6**, 2808 (1972); **6**, 2838 (1972); K. G. Chakraborty, Phys. Rev. B **10**, 2197 (1977); M. G. Cottam, *ibid.* **10**, 2383 (1977); R. Pirc and P. Prelovšek, Phys. Rev. B **15**, 4303 (1977); M. G. Cottam, D. R. Tilley, and B. Zeks, J. Phys. C **17**, 1793 (1984).

[8] F. Keffer, in *Handbook of Physics*, Vol. 18 (Springer, Berlin, 1966), p. 1.

[9] R. P. Singh, Z. C. Tao, and M. Singh, Phys. Rev. B **46**, 1244 (1992); R. P. Singh and M. Singh, Phys. Status Solidi B **169**, 571 (1992); R. P. Singh, *ibid.* **B 179**, 187 (1993).

[10] M. Kac, in *Statistical Physics, Phase Transitions, and Superfluidity*, Vol. 1, edited by M. Chretien, E. P. Gross, and S. Deser (Gordon and Breach, New York, 1968), p. 241.

[11] C. Kittel and H. Shore, Phys. Rev. **138**, A1165 (1965).

[12] H. Al-Wahsh, M. Urban, and A. Czachor, J. Magn. Magn. Mater. **185**, 144 (1998); A. Czachor, Physica B **311**, 56 (2002).

[13] A. Czachor and H. Al-Wahsh, Phys. Rev. B **63**, 064419 (2001).

[14] A. Kumar and A. K. Gupta, Phys. Rev. B **28**, 3968 (1983).

[15] G.-Z. Wei and A. Du, J. Phys.: Condens. Matter **5**, 1203 (1993).

[16] V. Yu. Irkhin, A. A. Katanin, and M. I. Katsnelson, Phys. Rev. B **60**, 1082 (1999).

[17] J. Lou, J. Dai, S. Qin, Z. Su, and L. Yu, Phys. Rev. B **62**, 8600 (2000).

[18] K. H. Lee and S. H. Liu, Phys. Rev. **159**, 390 (1967).

[19] F. Keffer and R. Loudon, J. Appl. Phys. **32**, 25 (1961).

[20] A. Czachor and A. Holas, Phys. Rev. B **41**, 4674 (1990).

[21] V. Yu. Irkhin and A. A. Katanin, Phys. Rev. B **57**, 379 (1998).

[22] A. N. Rubtsov, J. Hlinka, and T. Janssen, Phys. Rev. E **61**, 126 (2000).