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Explanation of the Cu spin-wave excitation gap in Nd_2CuO_4

T. Pyttlik[1,a] and K.W. Becker[1]

Institut für Theoretische Physik, Technische Universität Dresden, 01062 Dresden, Germany

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Abstract. By use of a mean-field approach the spin-wave dispersion of the Cu degrees of freedom in the undoped high-T_C material Nd_2CuO_4 is investigated. The experimentally observed sharp decrease of the Cu spin-wave gap with increasing temperature in the range $T^* < T < T_N$ is explained by a paramagnetic-like susceptibility of the Nd spins which couple to the Cu subsystem. The degeneracy of the "in-plane" and "out-of-plane" polarized Cu spin-wave branches is shown to be lifted by the uniaxial anisotropy of the Cu-Cu nearest-neighbor interaction.

PACS. 74.72.Dn La-based cuprates – 75.30.Cr Saturation moments and magnetic susceptibilities – 75.40.Gb Dynamic properties (dynamic susceptibility, spin waves, spin diffusion, dynamic scalling, etc.)

1 Introduction

Since the discovery of electron superconductivity [1] and heavy fermion-like behavior [2] in Ce-doped Nd_2CuO_4, its magnetic structure and magnetic excitations have been studied intensively. The magnetic structure of the undoped material is formed by Cu^{2+} spins and Nd^{3+} pseudospins. The latter are carried by the lowest crystalline electric field (CEF) doublet of the Nd^{3+} ions.

The Cu spins order in a noncollinear structure of antiferromagnetically ordered planes at a Néel temperature $T_N \approx 250$–280 K [3–7]. Below T_N, the Nd spins are also ordered due to strong interactions between Cu and Nd spins [8]. Two spin reorientation transitions were observed at 30 K and 75 K [4,5], where adjacent three-plane units consisting of a Cu plane and two Nd planes are rotated relatively to one another. Recently, a model has been proposed which explains this behavior by competition between different inter-unit interactions [9]. We will use this model, as well as an alternative model, as a starting point for our mean-field calculation.

The magnetic excitation spectrum of Nd_2CuO_4 has been attributed for low energies to Nd excitations and for high energies to Cu excitations. Inelastic neutron scattering experiments on polycristalline samples and single crystals at temperatures between 50 mK and 4 K were performed at low energies ($E < 0.8$ meV) [10–14]. The high energy ($E > 2.0$ meV) Cu spin-wave excitations have been studied by neutron scattering in single crystals at temperatures between 3–100 K [15–17]. Two different energy gaps were found and have been attributed to the "in-plane" (IP) and "out-of-plane" (OP) polarized Cu magnon branches. The gap energies drop sharply from 10–15 meV below 5 K to about 2 meV (IP) and 5 meV (OP) at 90 K, according to an approximate $(1/\sqrt{T})$-law. It was suggested that this temperature dependence should be related to a $(1/T)$-dependence of the polarization of the Nd ions in the molecular field generated by the antiferromagnetically ordered Cu spins [8,17,18]. A theoretical description of the spin-wave spectrum of the noncollinear Cu sublattice had already preceded these experimental results [19]. This work also adressed the important question why the Cu system orders in a noncollinear structure below T_C.

In this paper, we present a mean-field approach to derive quantitative expressions for the temperature dependence of the Cu spin-wave gaps at intermediate temperatures ($T^* < T \ll T_N$). The lower temperature $T^* \approx 2$ K is of the order of the Nd-Nd exchange coupling, so that for $T > T^*$ the thermal energy exceeds the interaction between the Nd ions. The temperature T^* should not be mixed up with a Nd ordering temperature, because the magnetic order of the Nd ions below T_N is induced by the Cu-Nd exchange which acts as a staggered magnetic field at the Nd sites.

Very recently the magnetic excitations for the coupled Cu-Nd system were studied in the framework of a spin-wave theory for both the Cu and Nd spins [9]. There, also an extension to finite temperatures was sketched which leads to results equivalent to those derived below. However, although using the same model as this work, we believe that our approach is much more transparent and allows a better interpretation of the anomalous temperature dependence of the Cu spin excitation gaps. As we concentrate on the Cu spin degrees of freedom, we do not reproduce the full spectrum of modes given in reference [9].

The paper is organized as follows: in Section 2 the model will be presented. The dynamical susceptibility of

[a] e-mail: pyttlik@physik.tu-dresden.de

the Cu subsystem will be derived in Section 3 and will be used to calculate the dispersion relations of the Cu spin-wave modes. In Section 4, the resulting expressions for the Cu spin-wave gaps will be matched to experimental data in order to determine the model parameters. We will use mean-field theory throughout this paper. An alternative route to determine the Cu spin-wave dispersions would be based on projection technique [20–22].

2 Model

The low temperature magnetic structure of Nd_2CuO_4 [3–8] is shown in Figures 1 and 2. The localized magnetic moments are supposed to interact *via* a Heisenberg exchange mechanism. For a better understanding of our subsequent calculations, the model used in reference [9] shall be presented briefly. Motivated by the observation that at the spin reorientation transitions only the relative orientation of adjacent three-plane units – but not the spin structure within these units – changes, the Cu-Nd interaction between nearest neighbors has been assumed to be the dominant interaction between Cu and Nd ions. The lattice is then decomposed into weakly interacting three-plane units consisting of a Cu plane sandwiched between two Nd planes as shown in Figure 1. We will consider only one of these units and completely neglect interactions between units. This reduces the three-dimensional crystal to a two-dimensional (planar) model system.

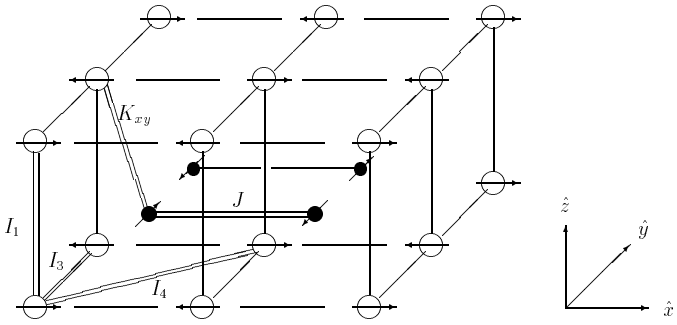

Fig. 1. Part of a three-plane unit comprising a Cu plane (full circles) and the adjacent Nd planes (open circles). Low temperature magnetic moments are indicated by arrows. The exchange parameters are marked by double lines connecting the corresponding lattice sites. The nearest-neighbor Cu-Nd interaction K_{xy} is of pseudodipolar type.

The interaction Hamiltonian

$$H^{int} = H^{\mathrm{Cu-Cu}} + H^{\mathrm{Nd-Nd}} + H^{\mathrm{Cu-Nd}} \tag{1}$$

is composed of three parts describing interactions between different species of ions. Both $H^{\mathrm{Cu-Cu}}$ and $H^{\mathrm{Nd-Nd}}$ have been used before to describe the respective isolated Cu and Nd spin systems in three dimensions [5,13,14,16,19]. However, for the investigation of the Cu spin-wave gaps it turns out to be necessary to treat the full Hamiltonian including the Cu-Nd interaction. The crystalline electric field Hamiltonian H^{CEF} for the Nd ions will not be explicitly included. It is assumed that all Nd ions are in the CEF groundstate because the next higher CEF doublet has an energy of 15.9 meV [23] and may be neglected for temperatures $T < 100$ K.

Denoting the Cu spins by $\mathbf{S}_i$ and the Nd pseudospins by $\mathbf{F}_{\pm,i}$, the Hamiltonian for the planar model is given by

$$H^{\mathrm{Cu-Cu}} = -J \sum_{\langle ij \rangle} \left(\mathbf{S}_i \cdot \mathbf{S}_j - \varepsilon S_i^z S_j^z \right) , \tag{2}$$

$$\begin{aligned} H^{\mathrm{Nd-Nd}} = &- \sum_i \mathbf{F}_{-,i} \cdot \underline{\underline{I_1}} \cdot \mathbf{F}_{+,i} \\ &- \sum_{\langle ij \rangle} \sum_\alpha \mathbf{F}_{\alpha,i} \cdot \underline{\underline{I_3}} \cdot \mathbf{F}_{\alpha,j} \\ &- \sum_{\langle\langle ij \rangle\rangle} \sum_\alpha \mathbf{F}_{\alpha,i} \cdot \underline{\underline{I_4}} \cdot \mathbf{F}_{\alpha,j} , \end{aligned} \tag{3}$$

$$H^{\mathrm{Cu-Nd}} = K_{xy} \sum_{[ij]} \sum_\alpha \left(\mathbf{S}_i^x \mathbf{F}_{\alpha,j}^y + \mathbf{S}_i^y \mathbf{F}_{\alpha,j}^x \right) \varphi_{ij} . \tag{4}$$

Here the first lower index $\alpha = +(-)$ on the Nd spins indicates spins lying on the upper (lower) Nd plane of the three-plane unit (*cf.* Fig. 1). The indices i or j denote the xy coordinates within the planes. The notations $\langle ij \rangle$ and $\langle\langle ij \rangle\rangle$ in equations (2, 3) refer to pairs of in-plane nearest and next-nearest neighbors respectively, while $[ij]$ in equation (4) refers to nearest-neighbor pairs on different planes. The orientation of the lattice has been chosen in a way that in the groundstate the Cu spins point in the $\pm y$ direction and the Nd spins in the $\pm x$ direction.

As an isotropic interaction between nearest neighbors on adjacent planes would vanish on the average due to the antiferromagnetic geometry of the groundstate (Fig. 2), the Cu-Nd interaction is assumed to be of pseudodipolar type [9]. The phase factor $\varphi_{ij} = e^{i\mathbf{Q}\cdot\mathbf{R}_i} e^{i\mathbf{Q}\cdot\mathbf{R}_{\alpha,j}}$ in equation (4) incorporates the directional dependence of this coupling. Here, $\mathbf{Q} = (\pi, \pi)$ is the antiferromagnetic wave vector and $\mathbf{R}_i$, $\mathbf{R}_{\alpha,j}$ denote lattice vectors.

The antiferromagnetic nearest-neighbor Cu-Cu coupling $J < 0$ has been found to be the dominant spin–spin interaction in Nd_2CuO_4 [5,24]. It is well known for the lamellar cuprates that this interaction is axially anisotropic as expressed by the dimensionless anisotropy parameter $\varepsilon > 0$ [5,16]. The anisotropy of the Cu-Cu exchange is the reason for the different gap energies of IP and OP polarized Cu spin waves. A next-nearest-neighbor interaction between Cu spins is presumably weaker than J and has not been included.

The Nd-Nd exchange tensors $\underline{\underline{I_\nu}}$ ($\nu = 1$–4) [13,14] are diagonal with $\underline{\underline{I_\nu}}^{xx} = \underline{\underline{I_\nu}}^{yy} = I_\nu^\perp < 0$, $\underline{\underline{I_\nu}}^{zz} = I_\nu^\parallel < 0$. Note that an additional nearest-neighbor exchange $\underline{\underline{I_2}}$ between Nd spins of adjacent planes belonging to different three-plane units will not be considered. Such an interaction would vanish within the mean-field approach used below.

The groundstate of the planar model is twofold degenerate, because the energy does not change if all spins are rotated by 180°. An additional twofold degeneracy arises from the fact that there are two types of three-plane units with Cu spins pointing in y direction or x direction.

Note that the part of the Hamiltonian acting on the Cu spins is not rotationally invariant due to the angular dependence of K_{xy} and the small anisotropy parameter ε. The effect of nonzero K_{xy} and ε on the isotropic system is to select the classical zero temperature groundstate shown in Figure 1 from among similar states with all spins rotated around a common axis. A finite $\varepsilon > 0$ makes states with Cu spins pointing in the z direction energetically less favorable than states with Cu spins pointing perpendicular to the z direction, while $K_{xy} > 0$ stabilizes the noncollinear arrangement of Cu and Nd spins in adjacent planes.

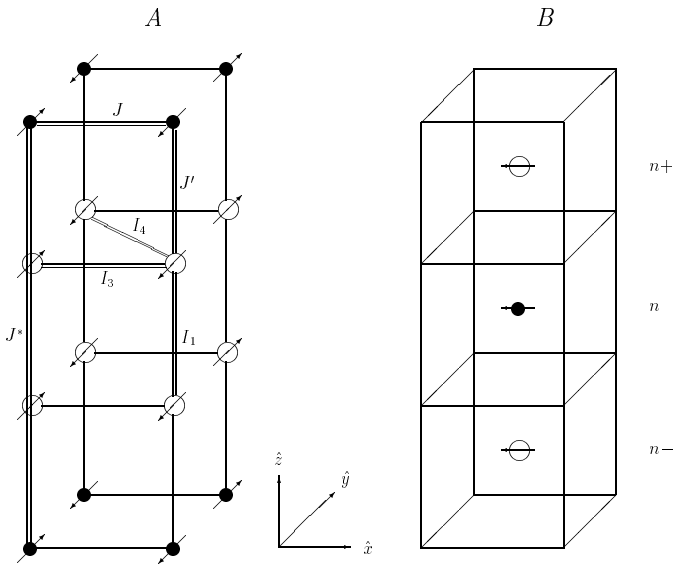

Fig. 2. Tetragonal unit cell of Nd_2CuO_4 with noncollinear sublattices A (left) and B (right) pulled apart. Labels on the right of the B-cell indicate vertical plane numbering. The coupling J^* on the left denotes an effective interaction between Cu z axis neighbors.

Alternatively, the lattice may be decomposed into the two noncollinear sublattices A and B (Fig. 2), in each of which the Cu and Nd ions are located on sites of a simple tetragonal lattice. Sublattice B is generated from sublattice A by translation by $(a/2, a/2, c/2)$ plus a 90°-rotation around the z axis. Any spin located on sublattice A is in a symmetrical position with respect to the antiferromagnetically ordered spins on sublattice B. Therefore, if only isotropic exchange is considered the interaction between both sublattices vanishes on the average, which motivates the decomposition of the lattice into the approximately noninteracting sublattices. The isotropic antiferromagnetic Cu-Nd exchange $J' < 0$ between nearest neighbors along the z axis is then assumed to be the dominant Cu-Nd interaction. In order to explain the parallel orientation of the Cu spins along the z axis, an effective ferromagnetic Cu-Cu interplane interaction $J^* > 0$ has to be included. Note that this interaction has to be stronger than the competing effective Cu-Cu interaction mediated by J' and I_1 which itself would lead to an antiparallel orientation of Cu spins along the z axis. It is further necessary to break the tetragonal symmetry of the Hamiltonian by enhancing the y (x) component of the Cu-Nd interactions within sublattice A (B), so that instead of equation (4) the interactions

$$H_A^{\mathrm{Cu-Nd}} = -J' \sum_n \sum_i \left[\mathbf{S}_{n,i} \cdot \left(\mathbf{F}_{n-,i} + \mathbf{F}_{n+,i} \right) + \varepsilon' S^y_{n,i} \left(F^y_{n-,i} + F^y_{n+,i} \right) \right] , \quad (5)$$

$$H_B^{\mathrm{Cu-Nd}} = -J' \sum_n \sum_i \left[\mathbf{S}_{n,i} \cdot \left(\mathbf{F}_{n-,i} + \mathbf{F}_{n+,i} \right) + \varepsilon' S^x_{n,i} \left(F^x_{n-,i} + F^x_{n+,i} \right) \right] \quad (6)$$

are used, where the indices n, $n+$, and $n-$ denote a Cu plane and the two adjacent Nd-planes, all on the same sublattice. The small anisotropy parameter $\varepsilon' > 0$ favors states with spins in sublattice A (B) pointing in the y (x) direction and therefore stabilizes the noncollinear magnetic structure. An anisotropy of this kind has not been proposed so far. However, the parameter ε' enters the resulting Cu spin-wave dispersions only in the combination $J'(1 + \varepsilon')$.

The AB-sublattice model in fact retains three-dimensionality, but as the Cu-Cu interaction J^* along the z axis is much smaller than J, there is virtually no dispersion of the Cu spin waves in z direction. The groundstate of the AB-sublattice model is also fourfold degenerate, because all spins may be flipped by 180° independently on each of the two sublattices without change of energy. As the interaction between the noncollinear sublattices A and B has been neglected, the AB-sublattice model explains neither the noncollinear structure nor the spin reorientation transitions. However, the planar model and the AB-sublattice model lead to equivalent Cu spin-wave gaps, while the results for the Nd spin-wave modes depend crucially on the model used [9,25].

3 Dynamical copper spin susceptibility

In the following, we want to evaluate the Cu-Cu spin susceptibility within a mean-field treatment by using the model (2–4). We start from the Fourier-transformed interaction Hamiltonian

$$H^{\mathrm{Cu-Cu}} = -J \sum_{\mathbf{q}} \gamma_3(\mathbf{q}) \left(\mathbf{S}_{\mathbf{q}} \cdot \mathbf{S}_{-\mathbf{q}} - \varepsilon S^z_{\mathbf{q}} S^z_{-\mathbf{q}} \right) , \quad (7)$$

$$H^{\mathrm{Nd-Nd}} = -\sum_{\mathbf{q}} \mathbf{F}_{+,\mathbf{q}} \cdot \underline{\underline{I_1}} \cdot \mathbf{F}_{-,-\mathbf{q}} - \sum_{\mathbf{q}} \sum_{\alpha} \gamma_3(\mathbf{q})\, \mathbf{F}_{\alpha,\mathbf{q}} \cdot \underline{\underline{I_3}} \cdot \mathbf{F}_{\alpha,-\mathbf{q}} - \sum_{\mathbf{q}} \sum_{\alpha} \gamma_4(\mathbf{q})\, \mathbf{F}_{\alpha,\mathbf{q}} \cdot \underline{\underline{I_4}} \cdot \mathbf{F}_{\alpha,-\mathbf{q}} \,, \tag{8}$$

$$H^{\mathrm{Cu-Nd}} = K_{xy} \sum_{\mathbf{q}} \sum_{\alpha} \gamma_{pd}(\mathbf{q}) \mathbf{S}_{\mathbf{q}} \cdot \tilde{\mathbf{F}}_{\alpha,\mathbf{q}} \,, \tag{9}$$

where functions $\gamma_3(\mathbf{q}) = 2\big(\cos(q_x) + \cos(q_y)\big)$, $\gamma_4(\mathbf{q}) = 4\cos(q_x)\cos(q_y)$ and $\gamma_{pd}(\mathbf{q}) = 4\sin(q_x/2)\sin(q_y/2)$ have been introduced. Note that the angular dependence of the pseudodipolar interaction leads to sine functions in γ_{pd}, where cosines would be expected for an isotropic interaction. The vector $\tilde{\mathbf{F}}_{\alpha,\mathbf{q}} = \big(F^y_{\alpha,\mathbf{q}},\ F^x_{\alpha,\mathbf{q}},\ 0\big)$ in equation (9) is formed by interchanging the x and y component of the Nd spin $\mathbf{F}_{\alpha,\mathbf{q}}$ and setting the z component to zero.

In addition, a weak time- and space-dependent external magnetic field $\mathbf{H}e^{i\omega t}$ is coupled to the Cu spins by

$$H^Z(t) = g\mu_B \sum_{\mathbf{q}} \mathbf{S}_{\mathbf{q}} \cdot \mathbf{H}_{-\mathbf{q}} \cos(\omega t) \,, \tag{10}$$

so that the Hamiltonian now reads

$$H(t) = H^{int} + H^Z(t) \,. \tag{11}$$

The dynamical properties of the Cu excitations will be derived from the equation of motion

$$\frac{d}{dt}\Big\langle \mathbf{S}_{\mathbf{q}'}(t) \Big\rangle = -\frac{i}{\hbar}\Big\langle \left[\mathbf{S}_{\mathbf{q}'}(t),\ H(t)\right]_- \Big\rangle \,, \tag{12}$$

where the brackets $\big\langle \ldots \big\rangle$ denote dynamical expectation values. By use of the Hamiltonian (11), we obtain the expression

$$\begin{aligned}\frac{d}{dt}\Big\langle \mathbf{S}_{\mathbf{q}'} \Big\rangle &= g\mu_B \sum_{\mathbf{q}} \mathbf{H}_{\mathbf{q}}(t) \times \Big\langle \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle \\ &-2J \sum_{\mathbf{q}} \gamma_3(\mathbf{q}) \left(\Big\langle \mathbf{S}_{\mathbf{q}} \times \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle - \varepsilon \Big\langle S^z_{\mathbf{q}}\, \hat{\mathbf{z}} \times \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle \right) \\ &+K_{xy} \sum_{\mathbf{q}} \sum_{\alpha} \gamma_{pd}(\mathbf{q}) \Big\langle \tilde{\mathbf{F}}_{\alpha,\mathbf{q}} \times \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle \,.\end{aligned} \tag{13}$$

Next, the spin operators are decomposed into their static expectation values leading to molecular fields and into smaller fluctuation parts

$$\mathbf{S}_{\mathbf{q}} = \langle \mathbf{S}_{\mathbf{q}} \rangle_0 + \delta \mathbf{S}_{\mathbf{q}} \,, \tag{14}$$

$$\mathbf{F}_{\alpha,\mathbf{q}} = \langle \mathbf{F}_{\alpha,\mathbf{q}} \rangle_0 + \delta \mathbf{F}_{\alpha,\mathbf{q}} \,. \tag{15}$$

Subsequently, all fluctuation terms of quadratic order will be neglected, for example

$$\begin{aligned}\Big\langle \mathbf{S}_{\mathbf{q}} \times \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle &\approx \Big\langle \mathbf{S}_{\mathbf{q}} \Big\rangle_0 \times \Big\langle \delta \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle \\ &+ \Big\langle \delta \mathbf{S}_{\mathbf{q}} \Big\rangle \times \Big\langle \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle_0 + \Big\langle \mathbf{S}_{\mathbf{q}} \Big\rangle_0 \times \Big\langle \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle_0 \,.\end{aligned} \tag{16}$$

At temperatures $T \ll T_N$, due to the antiferromagnetic order of the Cu spins all static expectation values $\langle \mathbf{S}_{\mathbf{q}} \rangle_0$ vanish except those with wave vector $\mathbf{Q} = (\pi, \pi)$. Furthermore, the Cu spins are assumed to be oriented parallel to the y direction, so that $\langle \mathbf{S}_{\mathbf{Q}} \rangle_0$ will be replaced by $S_{\mathbf{Q}}\, \hat{\mathbf{y}}$, where $\hat{\mathbf{y}}$ is a unit vector and $S_{\mathbf{Q}}$ represents the staggered moment per Cu site. Consequently, all terms of quadratic order in the static Cu spin expectation values vanish due to the vector products (*cf.* Eq. (16)). The equation of motion then reduces to

$$\begin{aligned}\frac{d}{dt}\Big\langle \delta \mathbf{S}_{\mathbf{q}'} \Big\rangle &= S_{\mathbf{Q}}\, \hat{\mathbf{y}} \times \Big[-g\mu_B\, \mathbf{H}_{\mathbf{q}'-\mathbf{Q}}(t) \\ &+2J\Big(\gamma_3(\mathbf{q}'-\mathbf{Q}) - \gamma_3(\mathbf{Q})\Big) \Big\langle \delta \mathbf{S}_{\mathbf{q}'-\mathbf{Q}} \Big\rangle \\ &-2\varepsilon J \gamma_3(\mathbf{q}'-\mathbf{Q}) \Big\langle \delta S^z_{\mathbf{q}'-\mathbf{Q}} \Big\rangle \hat{\mathbf{z}} \\ &-K_{xy} \sum_{\alpha} \gamma_{pd}(\mathbf{q}'-\mathbf{Q}) \left(\Big\langle \tilde{\mathbf{F}}_{\alpha,\mathbf{q}'-\mathbf{Q}} \Big\rangle_0 + \Big\langle \delta \tilde{\mathbf{F}}_{\alpha,\mathbf{q}'-\mathbf{Q}} \Big\rangle \right) \Big] \\ &+K_{xy} \sum_{\mathbf{q}} \sum_{\alpha} \gamma_{pd}(\mathbf{q}) \Big\langle \tilde{\mathbf{F}}_{\alpha,\mathbf{q}} \Big\rangle_0 \times \Big\langle \delta \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle \,.\end{aligned} \tag{17}$$

As the external magnetic field $\mathbf{H}_{\mathbf{q}}$ was assumed to be weak, the fluctuation part of $\Big\langle \mathbf{S}_{\mathbf{q}'-\mathbf{q}} \Big\rangle$ has been omitted in the Zeeman term.

In order to eliminate the remaining Nd spin operators, linear response theory will be used. In an adiabatic approximation the dynamical expectation values $\Big\langle \delta \tilde{\mathbf{F}}_{\alpha,\mathbf{q}'-\mathbf{Q}} \Big\rangle$ in equation (17) can be neglected. This is reasonable because we are only interested in Cu spin wave excitations which have much higher energies than the Nd spin waves. Therefore, the time-dependent contributions $\Big\langle \delta \tilde{\mathbf{F}}_{\alpha,\mathbf{q}'-\mathbf{Q}} \Big\rangle$ are negligibly small. Furthermore, the Nd expectation values are independent of the position of the Nd spin above or below the Cu plane, so that the sums over $\alpha = +, -$ result in a factor of 2. The static expectation value $\Big\langle \tilde{\mathbf{F}}_{\mathbf{q}'-\mathbf{Q}} \Big\rangle_0$ will be evaluated within a mean-field approach. The Nd subsystem is subject to a molecular field $\mathbf{h}_{\mathrm{Cu}}(\mathbf{q})$ generated by the Cu moments. Up to first order in the Cu-Nd exchange the static response of the Nd moments is given by

$$\Big\langle \mathbf{F}_{\mathbf{q}} \Big\rangle_0 = \underline{\underline{\chi}}^{\mathrm{Nd}}_0(\mathbf{q}) \cdot \mathbf{h}_{\mathrm{Cu}}(\mathbf{q}) \,. \tag{18}$$

Here $\underline{\underline{\chi}}^{\mathrm{Nd}}_0(\mathbf{q})$ is the static susceptibility matrix of the pure Nd system whereas $\mathbf{h}_{\mathrm{Cu}}(\mathbf{q})$ acts as a staggered field in x direction

$$\mathbf{h}_{\mathrm{Cu}}(\mathbf{q}) = h^x_{\mathrm{Cu}}(\mathbf{Q})\, \delta_{\mathbf{q}\mathbf{Q}}\, \hat{\mathbf{x}} \,, \tag{19}$$

$$h^x_{\mathrm{Cu}}(\mathbf{Q}) = -4K_{xy} \Big\langle S^y_{\mathbf{Q}} \Big\rangle_0 = -4K_{xy} S_{\mathbf{Q}} \,. \tag{20}$$

At temperatures $T > T^*$ where T^* is a temperature of the order of the Nd-Nd interactions, the Nd susceptibility has a paramagnetic-like shape. One finds that $\underline{\underline{\chi}}^{\mathrm{Nd}}_0(\mathbf{q})$ is diagonal with $\underline{\underline{\chi}}^{xx\,\mathrm{Nd}}_0(\mathbf{q}) = \underline{\underline{\chi}}^{yy\,\mathrm{Nd}}_0(\mathbf{q}) = \chi^{\perp\,\mathrm{Nd}}_0(\mathbf{q})$ and

$\underline{\underline{\chi}}_0^{zz\,\mathrm{Nd}}(\mathbf{q}) = \chi_0^{\parallel\,\mathrm{Nd}}(\mathbf{q})$, where for $\eta = \perp, \parallel$

$$\chi_0^{\eta\,\mathrm{Nd}}(\mathbf{q}) = \left(\frac{k_B T}{F^2} - \Big(I_1^\eta + 2I_3^\eta\gamma_3(\mathbf{q}) + 2I_4^\eta\gamma_4(\mathbf{q})\Big)\right)^{-1} . \quad (21)$$

Here $F = 1/2$ denotes the pseudospin of the lowest Nd CEF doublet. In evaluating the $\mathbf{q}$-dependent part of equation (21), the parameter values $\underline{\underline{I_\nu}}$ (ν = 1–4) given by Henggeler *et al.* [13,14] for the exchange interactions of the Nd pseudospins will be used. Equation (18) is used to eliminate $\left\langle \tilde{\mathbf{F}}_{\mathbf{q}'-\mathbf{q}} \right\rangle_0 = \left\langle F^x_{\mathbf{q}'-\mathbf{q}} \right\rangle_0 \hat{\mathbf{y}}$ from equation (17), so that

$$\frac{d}{dt}\left\langle \delta\mathbf{S}_\mathbf{q} \right\rangle = S_\mathbf{Q}\,\hat{\mathbf{y}} \times \Bigg[- g\mu_B \mathbf{H}_{\mathbf{q}-\mathbf{Q}}(t) + \tilde{J}(\mathbf{q}-\mathbf{Q})\left\langle \delta\mathbf{S}_{\mathbf{q}-\mathbf{Q}} \right\rangle - 2\varepsilon J\gamma_3(\mathbf{q}-\mathbf{Q})\left\langle \delta S^z_{\mathbf{q}-\mathbf{Q}} \right\rangle \hat{\mathbf{z}} \Bigg] , \quad (22)$$

where

$$\tilde{J}(\mathbf{q}) = 2J\Big(\gamma_3(\mathbf{q}) - \gamma_3(\mathbf{Q})\Big) - 8K^2_{xy}\gamma_{pd}(\mathbf{Q})\chi_0^{\perp\,\mathrm{Nd}}(\mathbf{Q}) . \quad (23)$$

As is obvious from equation (22), fluctuations of the longitudinal component $S^y_\mathbf{q}$ vanish. The fluctuations of the x component constitute the IP polarized spin-wave modes, those of the z component the OP modes. Equation (22) forms two sets of coupled linear differential equations for the two sets of variables

$$\left\{ \left\langle \delta S^x_\mathbf{q} \right\rangle , \left\langle \delta S^z_{\mathbf{q}-\mathbf{Q}} \right\rangle \right\} , \left\{ \left\langle \delta S^z_\mathbf{q} \right\rangle , \left\langle \delta S^x_{\mathbf{q}-\mathbf{Q}} \right\rangle \right\} . \quad (24)$$

From these sets one can obtain the dynamical susceptibility $\underline{\underline{\chi}}^{\mathrm{Cu}}(\mathbf{q},\omega)$, *e.g.*, for the first set in (24)

$$\begin{pmatrix} \left\langle \delta S^x_\mathbf{q} \right\rangle \\ \left\langle \delta S^z_{\mathbf{q}-\mathbf{Q}} \right\rangle \end{pmatrix} = g\mu_B Re \left\{ \underline{\underline{\chi}}^{\mathrm{Cu}}(\mathbf{q},\omega) \cdot \begin{pmatrix} H^x_\mathbf{q} e^{i\omega t} \\ H^z_{\mathbf{q}-\mathbf{Q}} e^{i\omega t} \end{pmatrix} \right\} , \quad (25)$$

where

$$\underline{\underline{\chi}}^{\mathrm{Cu}}(\mathbf{q},\omega) = \frac{S_\mathbf{Q}}{\omega^2(\mathbf{q}) - \omega^2} \times \begin{pmatrix} \Big(\tilde{J}(\mathbf{q}-\mathbf{Q}) - 2\varepsilon J\gamma_3(\mathbf{q}-\mathbf{Q})\Big)S_\mathbf{Q} & -i\omega \\ i\omega & \tilde{J}(\mathbf{q})S_\mathbf{Q} \end{pmatrix} . \quad (26)$$

The dispersion relation $\omega(\mathbf{q})$ for the Cu spin-wave excitations is found from the poles of the susceptibility

$$\omega^2_{IP}(\mathbf{q}) = \Big(\tilde{J}(\mathbf{q}-\mathbf{Q}) - 2\varepsilon J\gamma_3(\mathbf{q}-\mathbf{Q})\Big)\,\tilde{J}(\mathbf{q})S^2_\mathbf{Q} . \quad (27)$$

The second set of variables in (24) yields

$$\omega^2_{OP}(\mathbf{q}) = \Big(\tilde{J}(\mathbf{q}) - 2\varepsilon J\gamma_3(\mathbf{q})\Big)\,\tilde{J}(\mathbf{q}-\mathbf{Q})S^2_\mathbf{Q} , \quad (28)$$

which is different from equation (27) due to the appearance of the anisotropy ε. By reference to the equation of motion (22) the corresponding eigenmodes have been identified as IP and OP polarized excitations, respectively. The derivation of the excitation energies (27, 28) is also possible within the framework of projection formalism [20–22].

The dispersions are plotted in Figure 3, where the parameter values given in Section 4 have been used. In the Γ-M direction our results are in qualitative agreement with those of reference [19], except that we find a gap at the Γ-point. The mode energies decrease significantly with increasing temperature. The energy difference $\Delta\omega(\mathbf{q}) = \omega_{OP}(\mathbf{q}) - \omega_{IP}(\mathbf{q})$ of the two polarization branches is most pronounced at the antiferromagnetic Bragg point $X_C = (\pi,\pi)$ and at the center point $\Gamma = (0,0)$, as is shown in Figure 4. At the point $M = (\pi, 0)$ the two Cu spin-wave modes are degenerate.

Note that an analogous calculation can also be performed in the three-dimensional AB-sublattice model by use of the Cu-Nd interactions (5, 6). In this case, the Cu exchange field (Eq. (20)) has to be replaced by

$$h^y_{\mathrm{Cu}}(\mathbf{Q}) = J'(1+\varepsilon')S_\mathbf{Q} \quad (29)$$

for the A sublattice and an equivalent expression for the B sublattice. This leads to an overall replacement of $16K^2_{xy}$ by $J'^2(1+\varepsilon')^2$ without further change of the structure of the equations. Therefore, with respect to the Cu excitations, the planar model and the AB-sublattice model are equivalent.

4 Discussion

The excitation gaps Δ_{IP} and Δ_{OP} are obtained by evaluating equations (27, 28) at $\mathbf{q} = \mathbf{Q}$

$$\Delta^2_{IP} = -32K^2_{xy}\chi_0^{\perp\,\mathrm{Nd}}(\mathbf{Q})\left(\tilde{J}(\mathbf{0}) - 8\varepsilon J\right)S^2_\mathbf{Q} , \quad (30)$$

$$\Delta^2_{OP} = \Delta^2_{IP} + \Delta^2 , \quad (31)$$

where

$$\Delta^2 = 8\varepsilon J\left(\tilde{J}(\mathbf{0}) + \tilde{J}(\mathbf{Q})\right)S^2_\mathbf{Q} . \quad (32)$$

The empirical relation $\Delta^2_{OP} = \Delta^2_{IP} + \Delta^2$ proposed by Ivanov *et al.* [17] in discussing their experimental data is reproduced within our approach.

In evaluating the excitation energies, the temperature dependence of the staggered moment per Cu site

$$S_\mathbf{Q} = 0.606\left(1 - \frac{T}{T_N}\right)^\beta S \quad (33)$$

has been used [5–7,9,26,27]. $S = 1/2$ denotes the Cu spin. The exponent $\beta = 0.3$ has been determined for Nd_2CuO_4 [28], with other lamellar cuprates showing a similar temperature dependence. Under comparatively weak Cu-Nd coupling ($K_{xy} \ll |J|$) and weak external field the Cu subsystem is treated as a two-dimensional Heisenberg antiferromagnet [29,30], for which quantum fluctuations reduce

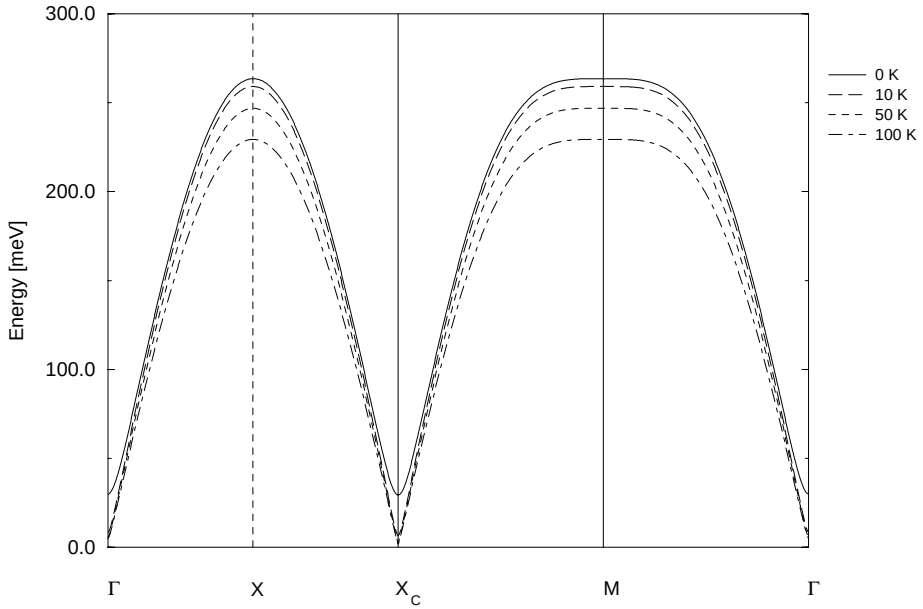

Fig. 3. Dispersion of the Cu spin-wave excitations in the tetragonal ΓMX_C plane at temperatures $T = 0$, 10, 50, 100 K. Γ denotes the center of the Brillouin zone, X_C the antiferromagnetic Bragg point, and X is halfway between Γ and X_C [25]. The interaction parameters are those presented in Section 4. Note that the energy difference between IP and OP polarized excitations is much smaller than the energy scale of this plot.

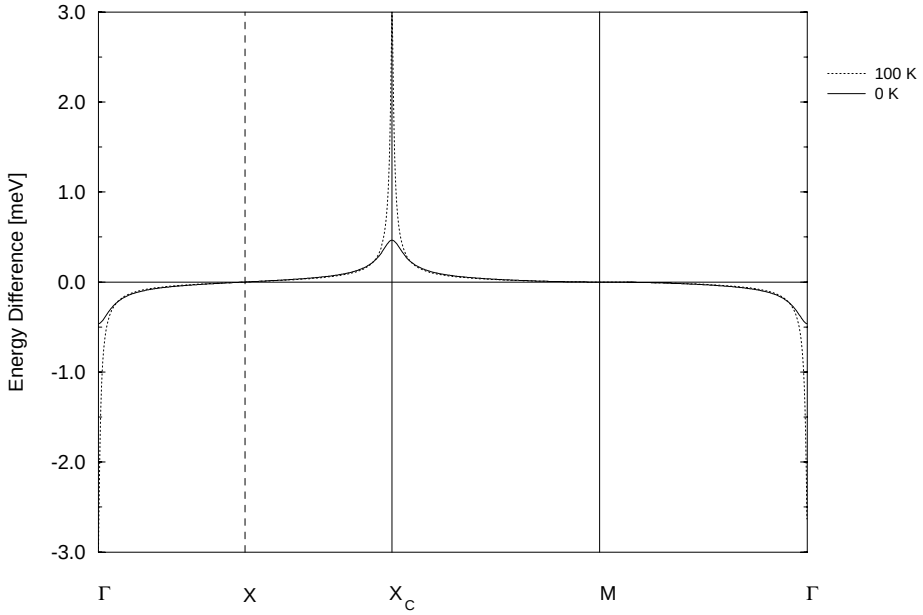

Fig. 4. Energy difference $\Delta\omega(\mathbf{q}) = \omega_{OP}(\mathbf{q}) - \omega_{IP}(\mathbf{q})$ between Cu spin-wave modes in the tetragonal ΓMX_C plane at temperatures $T = 0$, 100 K. All parameters are those used in Figure 3. Note the difference in scale as compared to Figure 3.

the staggered moment at $T = 0$ by a factor ~ 0.606 compared to the two-dimensional Ising antiferromagnet. It has to be stressed that due to the occurence of $S_{\mathbf{Q}}$ in equation (32) the quantity Δ is not temperature-independent as the earlier notation ($\Delta_{OP}(T \to \infty)$, *cf.* Ref. [17]) might suggest, but rather vanishes for $T \to T_N$.

Below T_N the IP excitation shows a finite gap which vanishes if the anisotropic Cu-Nd interaction K_{xy} is switched off. In that case, zero-energy Goldstone modes occur to restore the tetragonal symmetry of the Hamiltonian which is broken by the magnetic structure of the groundstate. On the contrary, for $\varepsilon \neq 0$ the OP gap remains finite even if $K_{xy} = 0$. This is due to the fact that an OP polarized excitation cannot restore tetragonal symmetry. In the limit $\varepsilon = K_{xy} = 0$, where the Hamiltonian coupling to the Cu spins has full rotational symmetry, there are also OP polarized Goldstone modes and both gaps vanish.

To obtain quantitative results we used the interaction parameters of references [13,14,24]

$$J = -108 \pm 6 \text{ meV},$$
$$I_1^{\perp} = -0.43 \pm 0.03 \text{ meV},$$
$$I_3^{\perp} = -0.07 \pm 0.01 \text{ meV},$$
$$I_4^{\perp} = -0.04 \pm 0.01 \text{ meV}.$$

This leaves the parameters K_{xy} and ε to be determined. From a fit to the gap data we obtain

$$K_{xy} = 0.18 \pm 0.02 \text{ meV}, \quad (34)$$
$$\varepsilon = 2.0 \pm 0.2 \times 10^{-4} . \quad (35)$$

The magnitude of the anisotropy ε is in excellent agreement with literature values [5]. The synopsis of experimental data and our theoretical description is given in Figure 5. The sharp decrease of Δ_{IP} and Δ_{OP} with T is due to the strong temperature dependence of the paramagnetic susceptibility $\chi_0^{\perp\,\mathrm{Nd}}(\mathbf{q})$ which is of Curie-Weiss type (*cf.* Eq. (21)) for the pure Nd subsystem, *i.e.* without coupling to the Cu spins.

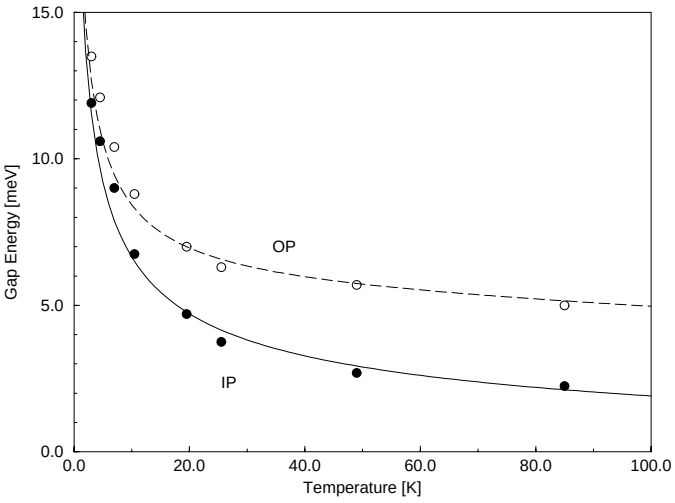

Fig. 5. Temperature dependence of energy gaps for "in-plane" (IP) and "out-of-plane" (OP) polarized Cu spin waves. Circles: experimental data from Ivanov *et al.* [17]; lines: modeled temperature dependence with parameters given in the text.

A description of Cu and Nd spin waves in Nd_2CuO_4 has already been performed by Sachidanandam *et al.* [9]. Contrary to our approach, they used spin-wave theory to obtain the Cu excitation energies at $T = 0$ and then generalized the dispersions to finite temperatures. Though their results are equivalent to ours, we believe that our approach is more transparent as it concentrates on the explanation of the temperature dependence of the Cu spin-wave gaps and the underlying physical ideas. Furthermore, we have shown that the resulting Cu spin-wave gaps are independent of the model used (three-plane unit or AB-sublattice model), as is checked most conveniently by projection formalism.

5 Conclusions

We have investigated the static and dynamical properties of the undoped high-T_C material Nd_2CuO_4 in the temperature range $T^* < T \ll T_N$ focusing on the high-energy Cu spin-wave excitations. In order to explain the observed temperature dependence of the Cu spin-wave gaps, we combined the interaction Hamiltonians of the isolated Cu and Nd subsystems with a third Hamiltonian describing anisotropic nearest-neighbor Cu-Nd exchange. We restricted our calculations to a single three-plane unit consisting of a Cu plane sandwiched between two Nd planes. Interactions between different three-plane units were completely neglected in this two-dimensional model. A three-dimensional model which decouples the two noncollinear sublattices of Nd_2CuO_4 was found to give equivalent results.

In the considered temperature range the static susceptibility $\underline{\underline{\chi}}_0^{\mathrm{Nd}}$ of the pure Nd subsystem without any coupling to the Cu spins shows a Curie-Weiss temperature dependence. Due to nonvanishing Cu-Nd exchange coupling, the molecular field of the antiferromagnetically ordered Cu spins induces a temperature-dependent ordered moment on the Nd sites which in turn acts back on the Cu spins and thus causes the dramatic increase of the Cu spin-wave gap below 30 K. The IP and OP polarized modes are nondegenerate due to a uniaxial anisotropy ε of the nearest-neighbor Cu-Cu exchange interaction.

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References

1. Y. Tokura, H. Takagi, S. Uchida, Nature **337**, 345 (1989).
2. T. Brugger, T. Schreiner, G. Roth, P. Adelmann, G. Czjzek, Phys. Rev. Lett. **71**, 2481 (1993).
3. Y. Endoh, M. Matsuda, K. Yamada, K. Kakurai, Y. Hidaka, G. Shirane, R.J. Birgeneau, Phys. Rev. B **40**, 7023 (1989).
4. S. Skanthakumar, H. Zhang, T.W. Clinton, W.-H. Li, J.W. Lynn, I.W. Sumarlin, Z. Fisk, S.-W. Choeng, Physica C **160**, 124 (1989).
5. M. Matsuda, K. Yamada, K. Kakurai, H. Kadowaki, T.R. Thurston, Y. Endoh, Phys. Rev. B **42**, 10098 (1990).
6. A.S. Cherny, E.N. Khats'ko, G. Chouteau, J.M. Louis, A.A. Stepanov, P. Wyder, S.N. Barilo, D.I. Zhigunov, Phys. Rev. B **45**, 12600 (1992).
7. S. Skanthakumar, J.W. Lynn, J.L. Peng, Z.Y. Li, Phys. Rev. B **47**, 6173 (1993).
8. J.P. Hill, A. Vigliante, D. Gibbs, J.L. Peng, R.L. Greene, Phys. Rev. B **52**, 6575 (1995).
9. R. Sachidanandam, T. Yildirim, A.B. Harris, A. Aharony, O. Entin-Wohlmann, Phys. Rev. B **56**, 260 (1997).
10. M. Loewenhaupt, P. Fabi, S. Horn, P. v. Aken, A. Severing, J. Mag. Mag. Mat. **140–144**, 1293 (1995).

11. M. Loewenhaupt, A. Metz, N.M. Pyka, D. McK. Paul, J. Martin, V.H.M. Duijn, J.J.M. Franse, H. Mutka, W. Schmidt, Ann. Phys. **5**, 197 (1996).
12. W. Henggeler, T. Chattopadhyay, B. Roessli, D.I. Zhigunov, S.N. Barilo, A. Furrer, Z. Phys. B **99**, 465 (1996).
13. W. Henggeler, T. Chattopadhyay, P. Thalmeier, P. Vorderwisch, A. Furrer, Europhys. Lett. **34**, 537 (1996).
14. W. Henggeler, T. Chattopadhyay, B. Roessli, P. Vorderwisch, P. Thalmeier, D.I. Zhigunov, S.N. Barilo, A. Furrer, Phys. Rev. B **55**, 1269 (1997).
15. P. Bourges, L. Boudarene, D. Petitgrand, P. Galez, Physica B **180–181**, 447 (1992).
16. P. Bourges, A.S. Ivanov, D. Petitgrand, J. Rossat-Mignod, L. Boudarene, Physica B **186–188**, 925 (1993).
17. A.S. Ivanov, P. Bourges, D. Petitgrand, J. Rossat-Mignod, Physica B **213–214**, 60 (1995).
18. H. Casalta, P. Bourges, D. Petitgrand, A.S. Ivanov, Solid State Commun. **100**, 683 (1996).
19. V.L. Sobolev, H.L. Huang, Yu.G. Pashkevich, M.M. Larionov, I.M. Vitebsky, V.A. Blinkin, Phys. Rev. B **49**, 1170 (1994).
20. H. Mori, Progr. Theor. Phys. **34**, 423 (1965).
21. R. Zwanzig, in: *Lectures in Theoretical Physics*, vol. 3 (New York: Interscience 1961).
22. D. Forster, *Hydrodynamic Fluctuations, Broken Symmetry, and Correlation Functions* (Reading, MA: Benjamin 1975).
23. A.T. Boothroyd, S.M. Doyle, D.McK. Paul, R. Osborn, Phys. Rev. B **45**, 10075 (1992).
24. P.E. Sulewski, P.A. Fleury, K.B. Lyons, S.-W. Cheong, Z. Fisk, Phys. Rev. B **41**, 225 (1990).
25. P. Thalmeier, Physica C **266**, 89 (1996).
26. D. Vaknin, S.K. Sinha, D.E. Moncton, D.C. Johnston, J.M. Newsam, C.R. Safinya, J.E. King Jr., Phys. Rev. Lett. **58**, 2802 (1987).
27. J.D. Jorgensen, H.-B. Schuttler, D.G. Hinks, D.W. Capone II, K. Zhang, M.B. Brodsky, D.J. Scalapino, Phys. Rev. Lett. **58**, 1024 (1987).
28. S. Skanthakumar, J.W. Lynn, J.L. Peng, Z.Y. Li, J. Mag. Mag. Mat. **104–107**, 519 (1992).
29. P.W. Anderson, Phys. Rev. **86**, 694 (1952).
30. M. Vojta, K.W. Becker, Phys. Rev. B **54**, 15483 (1996).