Localization of spin excitations and disruption of long-range order in weakly-doped La_2CuO_4

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We use a microscopic model to study the spin states of lightly (acceptor) doped La_2CuO_4. We investigate long-range indirect interactions between the spins of holes localized near acceptors of various types, interactions that are mediated by the exchange of virtual magnons in the antiferromagnetic CuO_2 planes. We show that although this interaction changes sign in the insulating phase, so that long-range spin order does not occur in a system of holes occupying random sites in the crystal, its presence is sufficient to effectively disrupt the long-range antiferromagnetic order in the system of Cu^{2+} spins. We suggest that the mechanism for this disruption is fluctuations in the magnetic anisotropy caused by random strains generated by the impurity ions, and construct the phase diagram of the system.

1. INTRODUCTION

It is now well-established that the common ancestor of the family of high-temperature superconductors (HTSC), the compound La_2CuO_4, is an antiferromagnet (AFM) with a layered structure consisting of magnetically ordered CuO_2 planes (see Fig. 1) with large intralayer and very small interlayer exchange interactions (EI).[1-3] These crystalline and magnetic features are common to the other HTSC as well, all of which exhibit AFM ordering in CuO_2 planes.[4-8] There are several equivalent, but not identical, CuO_2 planes, a prerequisite for an increased number of magnetic sublattices. Specifically, these sublattices are four in number in La_2CuO_4, $YBa_2Cu_3O_{6+x}$ (1-2-3 ceramic), La_2NiO_4, Nd_2CuO_4, etc. The Néel temperature T_N for $3D$ ordering is high in all the HTSC (~ 300 K for La_2CuO_4, Refs. 1, 4, 5, 8, 9; ~ 400 K for 1-2-3 when $x < 0.15$, Refs. 1, 4, 6, 10), but drops to zero rather quickly when impurities are introduced. Thus, in $La_{2-x}(Sr,Ba)_xCuO_4$, T_N is zero even for $x \approx 2 \cdot 10^{-2}$, which corresponds to an impurity concentration $c = x/2 \approx 1\%$ for one unit cell in the CuO_2. In 1-2-3 the suppression of AFM occurs at a somewhat higher concentration ($x \sim 0.4$); however, the same scenario applies, i.e., the gradual disappearance of long-range magnetic order, although for small x (when the centers appear primarily in the Cu–O chains and not in the CuO_2 planes) the creation of other spin structures is possible (including noncollinear structures[1,11,12]).

It is noteworthy that, although experiments indicate that the valence of copper in the CuO_2 planes is independent of doping (i.e., the spin at a site is preserved), data on how this disruption of order takes place and the structure of the resulting "paraphase" in HTSC are somewhat ambiguous. According to the majority of experimental papers, only the long-range order is missing, while short-range AFM correlations are reliably attested (see the review Ref. 1); however, the precise characteristics of these correlations have not yet been established. In certain other publications (e.g., Refs. 13, 14) it has been asserted that the original structure is preserved even in the metallic phase, with the sole difference that the magnitude of the local magnetic moment is greatly decreased while the AFM vector fluctuates strongly in space and time.

It is important that significant variations in the magnetic properties of the system precede (if we exclude a proposed narrow region of coexistence) the appearance of conductivity, which, e.g., in $La_{2-x}Sr_xCuO_4$ occurs only at a finite and somewhat higher value $c \sim 5\%$ (the well-known phase diagrams of HTSC show this unambiguously; see Ref. 1). In other words, the destruction of long-range AFM order takes place while the HTSC is still in its insulating phase, for which the carriers are localized. Without question, this fact must be reflected in any description of HTSC with extremely small acceptor content. In our opinion, it is of fundamental significance, since it allows us to "decouple" the problem of the appearance of conductivity (as a result of the insulator-metal transition) from that of the destruction of the magnetism.

Although in the overwhelming majority of theoretical papers the mechanisms proposed for the destruction of long-

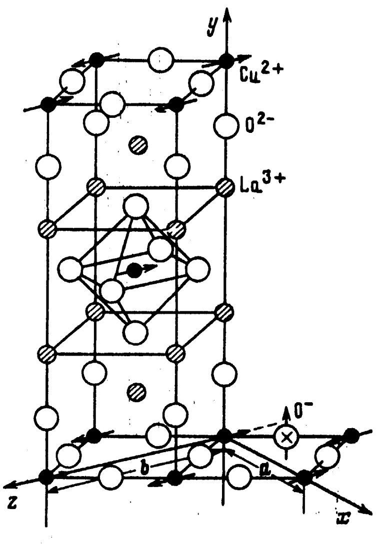

FIG. 1. Spin structure of a CuO_2 plane with localized hole.

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range AFM involve free-carrier models (the "spin bag" model,[15] field-theoretic models,[16,17] the Hubbard model,[17,18] etc.), recently more and more attention has been devoted to approaches that incorporate the localized character of carrier states at small concentrations. Let us begin by recalling the paper of Aharoni *et al.*[19] in which the concept of large ferromagnetic clusters in the CuO_2 planes which frustrate AFM order is introduced, along with later elaborations of these ideas.[20-22] However, in these papers the kinetic energy of a carrier near an impurity center was completely ignored. An attempt was made to include the kinetic energy in Ref. 23, where the Emery model was used to investigate suppression of AFM order within a finite cluster containing a mobile carrier. A deficiency of this approach is that it neglects *pp*-hopping, which probably plays a significant role by changing the character of the carrier motion.

In this paper we propose a model for describing the magnetic states of an impurity center while taking into account the real hierarchy of electronic interactions. We investigate centers in La_2CuO_4 originating from both the replacement of La^{3+} by Ba^{2+}, Sr^{2+}, and Ca^{2+} and the incorporation of above-stoichiometry levels of oxygen. We remind the reader that the experiments of Refs. 1 and 24 showed that both of these procedures lead to suppression of long-range AFM order in the insulating phase. In the new phase the spins of randomly-placed localized holes, by interacting with magnons of the host, are indirectly coupled. The character of this coupling allows us to infer certain things about the properties of the ground magnetic state of HTSC with a finite content of centers occupied by localized holes. However, as we will see below, for $c \sim 1\%$ the coupling cannot destroy the long-range AFM order (at least for temperatures that are not too low). Therefore, in this paper we propose another mechanism, which was invoked in a previous paper,[25] by which impurity centers can affect the magnetic order (including the value of T_N): fluctuations in the magnetic anisotropy of the CuO_2 planes due to random strains caused by impurities. We will show that for the values of strain observed in doped La_2CuO_4 this mechanism is completely realistic, and will attempt to construct the c–T phase diagram for this system theoretically.

2. ELECTRONIC STRUCTURE OF DOPING CENTERS

At this time, the available data indicate that excess holes exist primarily in the oxygen subsystem[1] and move within the $pd\sigma$ or $pd\pi$ bands. At low doping levels the holes are localized in the Coulomb potentials of the impurity ions. This localization is also facilitated by the well-known fact that in low-dimensional systems the appearance of a discrete level does not require any subsidiary conditions on the attractive potential.

We can estimate the radius r_0 of the corresponding electronic state by starting from the experimentally observed value of the activation energy[26] $\Delta\varepsilon \sim 10^{-1}$ eV of an impurity level and the width of the conduction band[27] $\Delta E_c \approx 2$ eV: $r_0 \sim |\mathbf{a}|(\Delta E_c/\Delta\varepsilon)^{1/2} \approx (2\text{–}4)|\mathbf{a}|$ (where $\mathbf{a}$ is the lattice constant vector). From this we see that effectively the carrier moves among a rather small number of O^{2-} ions close to the impurity center; in what follows we will limit this number by considering only those ions that lead to the lowest value of the Coulomb energy. In this case, to first approximation the splitting of the electronic energy within a cluster is determined by the kinetic characteristics (in particular the probability of *pp*-hopping), which are considerably larger than the magnetic interactions (i.e., the exchange interaction parameter). Therefore, as we will see below, the effect of a localized hole on the magnetic state of the crystal can be reduced to the problem of the interaction of the spin of a hole in its ground cluster state (with respect to kinetic energy) with the spins of the Cu^{2+}, or, what is the same thing, to the problem of an interstitial paramagnetic impurity.

It has often been noted in the literature that the distinctive feature of such an impurity center is its "frustrating" character, i.e., its symmetric placement with respect to antiparallel AFM vectors of the sublattices, which in the present case is due to the structure of the CuO_2 planes. Certain properties of a frustrating impurity in a host with the "easy axis" type of anisotropy have been studied previously[28] (see also Ref. 29). However, the anisotropy of magnetic interactions in HTSC is closer to the easy-plane type. Another distinguishing feature of HTSC, as we noted above, is the quasi-2D character of its magnetic properties, which allows us to limit our study to the CuO_2 planes.

It is noteworthy that, depending on the type of doping, centers of various kinds can form. In La_2CuO_{4+y} the above-stoichiometry oxygen ions occupy the positions (1/4, 1/4, 1/4) (Ref. 30), maximally perturbing the crystal field potential for a hole at the single O^{2-} ion in the next CuO_2 plane. If we assume that the excess holes are primarily localized by this type of ion, then an isolated "quadrupole," or "dumbbell," type of center (denoted db, see Fig. 2a) appears in the spin subsystem, with a C_2 axis of symmetry along one of the sides of the basal square, as is qualitatively described in Refs. 19, 20, and 22. However, if the system under discussion is $La_{2-x}(Ba,Sr,Ca)_xCuO_4$, then the impurity alkali-earth ions Me^{2+} are located near the centers of the 2D unit cells in the CuO_2 planes. In this case, the potential acting on a hole due to the simultaneous fields of the four O^{2-} is as small as possible, so that a different type of center (also frustrated) must appear, called a plaquette (denoted pl, see Fig. 2b)

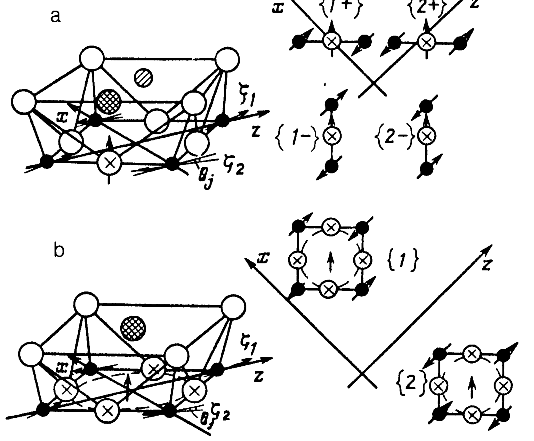

FIG. 2. Dumbbell (a) and plaquette (b) impurity centers in the La_2CuO_4 lattice and their possible types, taking into account both geometric and spin configurations (for nearest-neighbor host atoms). The doping impurity ions (O^{2-} in a, Sr^{2+} in b) are shown by double hatching.

with a C_4 axis of symmetry; this type of center was first described in Ref. 23. For each type of center there are various nonequivalent positions possible that differ in the spatial locations of the surrounding spins and the orientation of the symmetry axis C_2. For this last case, even when the localization radius of the hole is large, the point symmetry of the perturbation remains unchanged.

The lower symmetry centers (Fig. 2a) act as if they were part of more symmetric centers; therefore we will study them in detail, making the following simplification: the hole is trapped only by those O^{2-} ions for which the lowering of the Coulomb potential is a maximum, and cannot leave the plane in which it appears. Then the Hamiltonian for holes in the field of an impurity ion that forms a center of plaquette type near the **p**th CuO_2 cell, without including spin interactions, can be written in the form

$$\mathscr{H}_{\mathbf{p}} = \sum_{\alpha,\beta} T_{\mathbf{p}_\alpha \mathbf{p}_\beta} \overset{+}{a}_{\mathbf{p}_\alpha \sigma} a_{\mathbf{p}_\beta \sigma}, \tag{1}$$

where $T_{\mathbf{p}_\alpha \mathbf{p}_\beta}$ is the pp-hopping integral, and $a^+_{\mathbf{p}_\alpha \sigma}$ is the creation operator for a hole with spin σ on the αth O^{2-} ion of the **p**th cell. Taking into account the signs of the quantities $T_{\mathbf{p}_\alpha \mathbf{p}_\beta}$, which correspond to overlap of the nearest p-orbitals, it is easy to show that the four energy levels of the operator (1) equal $\varepsilon_1 = -2t$, $\varepsilon_2 = \varepsilon_3 = 0$, $\varepsilon_4 = 2t$ (where $t = |T|$). In the ground state, a localized hole naturally occupies the lowest level ε_1, whose corresponding cluster creation operator $c^+_{\mathbf{p}\sigma}$ is expressed in terms of the original "site" operators in the following way:

$$\overset{+}{c}_{\mathbf{p}\sigma} = {}^1/_2 (\overset{+}{a}_{\mathbf{p}_1\sigma} + \overset{+}{a}_{\mathbf{p}_2\sigma} - \overset{+}{a}_{\mathbf{p}_3\sigma} - \overset{+}{a}_{\mathbf{p}_4\sigma}). \tag{2}$$

The description of the low symmetry center of dumbbell type can be carried out in terms of the original operator $a^+_{\mathbf{p}_\alpha \sigma}$, since here the hole is localized on a single site.

3. SPECTRUM OF SPIN EXCITATIONS OF THE CRYSTAL AND THEIR PERTURBATIONS NEAR AN ISOLATED IMPURITY SPIN

In order to describe the spectrum of single-particle spin excitations of AFM La_2CuO_4 we will use the standard Hamiltonian which, applied to the CuO_2 plane, has the form[31]

$$\mathscr{H}_{\mathrm{AFM}} = \sum_{\mathbf{n}_1,\rho} [(J-\Delta J_t) S_{\mathbf{n}_1}{}^y S^y_{\mathbf{n}_1+\rho} + J(S_{\mathbf{n}_1}{}^x S^x_{\mathbf{n}_1+\rho} + S_{\mathbf{n}_1}{}^z S^z_{\mathbf{n}_1+\rho})$$
$$- D(S_{\mathbf{n}_1}{}^y S^z_{\mathbf{n}_1+\rho} - S_{\mathbf{n}_1}{}^z S^y_{\mathbf{n}_1+\rho})], \tag{3}$$

where $S_{\mathbf{n}_\alpha}$ is a spin operator for the **n**th Cu^{2+} ion of the αth magnetic sublattice ($\alpha = 1,2$), and J is the isotropic exchange interaction parameter between nearest neighbors; $\Delta J_t \ll J$ corresponds to the "easy plane" type of anisotropy in the tetragonal crystal structure; D is the parameter for the Dzyaloshinski (antisymmetric exchange) interaction, which in this case causes not only a departure of the spins from the basal xz plane but also a weak intraplanar rhombic anisotropy $\Delta J_{rh} \sim D^2/J < \Delta J_t$; and ρ is a vector that joins nearest-neighbor spins. After transforming the spin operators in (3), which are given in terms of their own (right-handed) coordinate systems with quantization axis ζ_α for the αth sublattice, to Holstein-Primakoff operators, isolating the part of the Hamiltonian that is quadratic in the latter, and carrying out the usual diagonalization procedures, we obtain the Hamiltonian

$$\mathscr{H}_m = \sum_{\mathbf{k},\mu} \Omega_\mu(\mathbf{k}) \beta_\mu{}^+(\mathbf{k}) \beta_\mu(\mathbf{k}), \tag{4}$$

where the eigenfrequencies of the magnon branches ($\mu = 1,2$) equal

$$\begin{gathered}
\Omega_\mu{}^2(\mathbf{k}) = [A_\mathbf{k} + (-1)^\mu B_\mathbf{k}]^2 - C_\mathbf{k}{}^2, \\
A_\mathbf{k} = (J\cos 2\theta + D\sin 2\theta + \Delta J_t \sin^2\theta) sz, \\
B_\mathbf{k} = [J\sin^2\theta + {}^1/_2(\Delta J_t \cos^2\theta - D\sin 2\theta)] sz\gamma_\mathbf{k}, \\
C_\mathbf{k} = [(J - {}^1/_2 \Delta J_t)\cos\theta + D\sin\theta] sz\gamma_\mathbf{k} \cos\theta, \\
\mathrm{tg}\, 2\theta = \frac{2D}{2J-\Delta J_t}, \qquad \gamma_\mathbf{k} = \frac{1}{z}\sum_\rho e^{i\mathbf{k}\rho}.
\end{gathered} \tag{5}$$

Here z is the number of nearest neighbors and $s = \langle S^{\zeta_\alpha}_{\mathbf{n}_\alpha} \rangle$; the Bose creation and annihilation operators for magnons are connected with the spin operators (to within linear terms) by the relations

$$S_{\mathbf{n}_\alpha}{}^x$$
$$= \frac{1}{2}\left(\frac{s}{N}\right)^{1/2} \sum_\mathbf{k} e^{i\mathbf{k}\mathbf{n}_\alpha} \{(-1)^\alpha (u_{1\mathbf{k}} + v_{1\mathbf{k}}) [\beta_1(\mathbf{k}) + \beta_1{}^+(-\mathbf{k})]$$
$$+ (u_{2\mathbf{k}} + v_{2\mathbf{k}}) [\beta_2(\mathbf{k}) + \beta_2^+(-\mathbf{k})], \tag{6}$$

$$S^{y}_{z\,n_\alpha} = \frac{1}{2i}\left(\frac{s}{N}\right)^{1/2} \sum_\mathbf{k} e^{i\mathbf{k}\mathbf{n}_\alpha} \{(-1)^\alpha (u_{1\mathbf{k}} - v_{1\mathbf{k}}) [\beta_1(\mathbf{k})$$
$$- \beta_1{}^+(-\mathbf{k})] + (u_{2\mathbf{k}} - v_{2\mathbf{k}})$$
$$\times [\beta_2(\mathbf{k}) - \beta_2{}^+(-\mathbf{k})]\} \begin{pmatrix} (-1)^\alpha \cos\theta \\ \sin\theta \end{pmatrix} - \frac{1}{2} \begin{pmatrix} -\sin\theta \\ (-1)^\alpha \cos\theta \end{pmatrix},$$

where

$$u_{\mu\mathbf{k}} = \left[\frac{A_\mathbf{k} + (-1)^\mu B_\mathbf{k} + \Omega_\mu(\mathbf{k})}{2\Omega_\mu(\mathbf{k})}\right]^{1/2}, \quad v_{\mu\mathbf{k}} = -(-1)^\mu (u_{\mu\mathbf{k}}{}^2 - 1)^{1/2}.$$

For small values of the two-dimensional wave vector with $\mathbf{ak} = [(ak_x)^2 + (bk_z)^2]^{1/2} \ll 1$, the dispersion law for both spin-wave branches takes the simple form:

$$\Omega_\mu{}^2(\mathbf{k}) = \Omega_{g\mu}{}^2 + J^2(\mathbf{ak})^2, \tag{7}$$

where $\Omega_{g1} = sz\sqrt{2J\Delta J_{rh}} = Dsz$ and $\Omega_{g2} = sz\sqrt{2J\Delta J_t}$ are the lower and upper energy gaps, respectively. In this case the stability of the long-range AFM order is determined by the quantity Ω_{g1}, and the Néel temperature T_N for such a quasi-$2D$ system can be found in the usual form (see, e.g., Ref. 32):

$$T_N = Jsz\left(\ln\frac{Jsz}{\Omega_{g1}}\right)^{-1} = Jsz\left(\ln\frac{J}{D}\right)^{-1} \tag{8}$$

It is natural, therefore, to expect that doping will affect the magnetic ordering of the host crystal most strongly by effectively decreasing the quantity Ω_{g1}. We note that an additional factor that stabilizes the long-range order in the quasi-$2D$ system is the interlayer exchange interaction, which causes dispersion of the magnons in directions transverse to the layers, an effect that is probably of the same order of magnitude as Ω_{g1}. However, for simplicity, in what follows we will neglect the existence of this dispersion, assuming that both stabilizing factors are suppressed by impurities simultaneously and in the same way.

The effect of an impurity center on the spin subsystem of an HTSC has a twofold character. On the one hand, impurity ions (both interstitial and substitutional) create rather strong local static distortions of the crystal lattice, as well as large local rotations $\varphi_0 \sim 30^\circ$ (compared to uniform values $\varphi \sim 4^\circ$ in the original crystal) of the oxygen octahedra closest to the impurities.[33] This leads to sizable local perturbations of the magnetic anisotropy parameters, and accordingly to a perturbation of the magnon spectrum. On the other hand, each of these centers creates a new spin degree of freedom in the system, associated with the spin carried by a hole in the crystal; consequently, new energy levels arise in the spectrum. We will consider these processes in the following sequence: first of all, we derive the spin Hamiltonian for holes localized near an isolated impurity center, and then use it to obtain an effective Hamiltonian for the spin interaction between holes at different centers. Then we determine the change in the static spin configuration and the magnetic anisotropy of the Cu^{2+} ions near an isolated center. Finally we find the change in the magnon spectrum and the ground state of the system as a whole.

In order to describe the spin interaction operators between the host and the carrier, we first give the Hamiltonian of an oxygen hole moving in the plane near the pth pl-impurity center (i.e., of plaquette type) and coupled to the spins of the Cu^{2+} ions. Taking into account terms of second order in the pd-transition (hybridization), we can reduce it[34,35] in the coordinate representation to the Hamiltonian of the Shubin-Vonsovsky polaron model:

$$\mathcal{H}_{pl} = \sum_{\mathbf{p}_\alpha \mathbf{p}_{\alpha 1} \mathbf{n}_\beta} \sum_{\sigma\sigma_1} \frac{\tilde{T}^{p\sigma,\,x^2-y^2}_{\mathbf{p}_\alpha \mathbf{n}_\beta} \tilde{T}^{x^2-y^2,\,p\sigma}_{\mathbf{n}_\beta \mathbf{p}_{\alpha 1}}}{\Delta E} a^+_{\mathbf{p}_\alpha \sigma} \hat{\boldsymbol{\sigma}}_{\sigma\sigma_1} a_{\mathbf{p}_{\alpha 1}\sigma_1} (\mathbf{S}_{\mathbf{n}_\beta})_{\sigma_1\sigma}, \tag{9}$$

where $\tilde{T}^{p_\sigma, x^2-y^2}_{\mathbf{p}_\alpha \mathbf{n}_\beta}$ is the matrix element for $pd\sigma$-hybridization between oxygen p-orbitals and x^2-y^2-states of the copper ion, ΔE is the difference in energy levels caused by the Hubbard repulsion of holes in the state Cu^{3+}, and $\hat{\sigma}$ is a vector of Pauli matrices that act in the space of the hole spin states. It is clear from (9) that processes can occur for which $\mathbf{p}_\alpha \neq \mathbf{p}_{\alpha 1}$; in this case we must assume that a copper ion $\mathbf{n}_\beta$ is close to the two oxygen ions between which the transition takes place. Then, taking into account the phases of the matrix elements $\tilde{T}$ for each pair of ions in a cluster (Fig. 2b) and also the relation (2), it is not difficult to reduce (9) to the form:

$$\mathcal{H}_{pl} = J' \sum_{\mathbf{p}_\alpha} \boldsymbol{\sigma}_{\mathbf{p}_\alpha} [\mathbf{S}_{(\mathbf{p}-\mathbf{a}/2)_\alpha} + \mathbf{S}_{(\mathbf{p}+\mathbf{a}/2)_\alpha} + \mathbf{S}_{(\mathbf{p}-\mathbf{b}/2)_\beta} + \mathbf{S}_{(\mathbf{p}+\mathbf{b}/2)_\beta}], \quad \alpha \neq \beta, \tag{10}$$

where the index α of the AFM sublattices labels one of the two possible configurations of an impurity pl-center, while the value of the constant $J' \approx |\tilde{T}|^2/\Delta E$ follows directly from (9) and (2). In an analogous fashion, we also obtain an expression for the Hamiltonian that describes the behavior of the spin of an oxygen hole $\boldsymbol{\sigma}_{\mathbf{p}_{\alpha\pm}} \equiv c^+_{\mathbf{p}_{\alpha\pm}\sigma} \hat{\boldsymbol{\sigma}}_{\sigma\sigma_1} c_{\mathbf{p}_{\alpha\pm}\sigma_1}$, localized at a low symmetry db-center with its symmetry axis along one side of the square (corresponding to the sign labels + or −):

$$\mathcal{H}_{ab} = J' \sum_{\mathbf{p}\alpha\pm} \boldsymbol{\sigma}_{\mathbf{p}_{\alpha\pm}} (\mathbf{S}_{[\mathbf{p}+(\mathbf{a}\pm\mathbf{b})/2]_\alpha} + \mathbf{S}_{[\mathbf{p}-(\mathbf{a}\pm\mathbf{b})/2]_\beta}), \quad \alpha \neq \beta. \tag{11}$$

It is clear from (11) (see also Fig. 2a) that for db impurities there are four nonequivalent configurations in all.

Using Eqs. (6), (10), and (11), we can write the general Hamiltonian for the spin interaction of localized holes with the AFM host as follows:

$$\mathcal{H}' = J' \sum_{\mathbf{p}_j, \boldsymbol{\rho}} \boldsymbol{\sigma}_{\mathbf{p}_j} \mathbf{S}_{\mathbf{p}_j + \boldsymbol{\rho}} = \sum_{\mathbf{p}_j} \Big\{ \omega_p \sigma^z_{\mathbf{p}_j} + N^{-1/2} \sum_{\mathbf{k},\mu} \exp(i\mathbf{k}\mathbf{p}_j) \sigma^-_{\mathbf{p}_j} \times [m^{(1)}_{j\mu}(\mathbf{k}) \beta_\mu(\mathbf{k}) + m^{(2)}_{j\mu}(\mathbf{k}) \beta^+_\mu(-\mathbf{k})] + \text{h.c.} \Big\}. \tag{12}$$

Here the label $j = \{1\}, \{2\}, \{1\pm\}$, and $\{2\pm\}$ enumerates all the possible (see Fig. 2) types and configurations of impurity centers. The eigenfrequencies $\omega_j = J'\sin\theta_j$ of these configurations are determined by the nonzero values of the molecular field generated by the host spins adjacent to the impurity, corresponding to departure of the jth spin (Fig. 2) from the basal plane by an angle θ_j (in the general case taking into account the influence of local strain $\theta_j \gg \theta$ of the lattice). Thus, the axis of quantization z of the impurity spin turns out to be perpendicular to this plane. The direction of the axis x is chosen to be the same for impurity spins and host spins, and in this case the constants for interaction of the fluctuations in the impurity spin with the magnons of the host equal

$$\begin{aligned} m^{(\nu)}_{\{j\pm\}1}(\mathbf{k}) &= i(-1)^j \frac{J' s^{1/2}}{2} \sin\left(\mathbf{k}\frac{\mathbf{a}\pm\mathbf{b}}{2}\right) [u_{1\mathbf{k}} + v_{1\mathbf{k}} + (-1)^\nu \sin\theta (u_{1\mathbf{k}} - v_{1\mathbf{k}})], \\ m^{(\nu)}_{\{j\pm\}2}(\mathbf{k}) &= \frac{J' s^{1/2}}{2} \cos\left(\mathbf{k}\frac{\mathbf{a}\pm\mathbf{b}}{2}\right) [u_{2\mathbf{k}} + v_{2\mathbf{k}} + (-1)^\nu \sin\theta (u_{2\mathbf{k}} - v_{2\mathbf{k}})] \end{aligned} \tag{13a}$$

for the dumbbell and

$$\begin{aligned} m^{(\nu)}_{\{j\}1}(\mathbf{k}) &= \frac{J' s^{1/2}}{2} \Big[(-1)^j \left(\cos\frac{\mathbf{ka}}{2} - \cos\frac{\mathbf{kb}}{2}\right)(u_{1\mathbf{k}} + v_{1\mathbf{k}}) - (-1)^\nu \sin\theta \left(\cos\frac{\mathbf{ka}}{2} + \cos\frac{\mathbf{kb}}{2}\right)(u_{1\mathbf{k}} - v_{1\mathbf{k}}) \Big], \\ m^{(\nu)}_{\{j\}2}(\mathbf{k}) &= \frac{J' s^{1/2}}{2} \Big[\left(\cos\frac{\mathbf{ka}}{2} + \cos\frac{\mathbf{kb}}{2}\right)(u_{2\mathbf{k}} + v_{2\mathbf{k}}) - (-1)^{\nu+j} \sin\theta \left(\cos\frac{\mathbf{ka}}{2} - \cos\frac{\mathbf{kb}}{2}\right)(u_{2\mathbf{k}} - v_{2\mathbf{k}}) \Big] \end{aligned} \tag{13b}$$

for the plaquette, where the label $\nu = 1, 2$ distinguishes the corresponding constants in the interaction operator (12).

The Hamiltonian (12) has a form that is characteristic of many problems in the theory of disordered spin systems (see, e.g., the review Ref. 29), as well as for insulating glasses containing two-level systems that interact with a strain field (see, e.g., Ref. 36). As will be clear below, anomalies in the magnetic properties of the subsystem of impurity spins and the crystal as a whole are determined in this case by the symmetries of the impurity centers and the dimensionality of the problem.

We note that in obtaining the Hamiltonian $\mathscr{H}'$ we have assumed that the interaction between the localized spin and the host spins is not strong enough for the spins to form a cluster, a possibility that was discussed, e.g., in Refs. 19–22. In these papers it was assumed that $J' \gg J$; since, first of all, the numerical calculations of Ref. 37 do not yield a strong inequality between J and J', and secondly, J' must be compared with the full exchange field Jz, we feel that clustering can be ignored. Finally, even when $J' \gg Jz$ holds, the resulting cluster of strongly coupled spins is located in zero mean field, so that its interactions with magnons reduces to expressions of the type (12).

4. EFFECTIVE INTERACTION BETWEEN IMPURITY SPINS

Using the full spin Hamiltonian of the crystal in the form $\mathscr{H} = \mathscr{H}_m + \mathscr{H}'$ [see (4) and (12)], by means of standard approaches to the theory of disordered systems[29,38] it is not difficult to find expressions for the Green's functions, e.g., $\langle\langle \beta_\mu(\mathbf{k}) | \beta_\mu^+(\mathbf{k}) \rangle\rangle_\omega$, whose poles are determined by the well-known dispersion equation

$$\omega^2 - \Omega_\mu{}^2(\mathbf{k}) - \mathrm{Re}\, R_\mu(\mathbf{k}, \omega^2) = 0. \tag{14}$$

Here, to first order in the concentration, the polarization operator $R_\mu(\mathbf{k},\omega^2)$ has the form

$$R_\mu(\mathbf{k}, \omega^2) = \frac{2\sigma}{N} \sum_{\mathbf{p}_j} \{\omega^2[|m_{j\mu}^{(1)}(\mathbf{k})|^2 - |m_{j\mu}^{(2)}(\mathbf{k})|^2] + \omega_j \Omega_\mu(\mathbf{k}) [|m_{j\mu}^{(1)}(\mathbf{k})|^2 + |m_{j\mu}^{(2)}(\mathbf{k})|^2]\} (\omega^2 - \omega_j^2)^{-1}, \tag{15}$$

where $\sigma = \langle \sigma_{p_j}^z \rangle$ is the mean value of the impurity spin, while the coupling constant is determined by Eqs. (13).

The value of the energy gap in the magnon spectrum when impurities are present equals $\tilde{\Omega}_{g1} = [\Omega_{g1}^2 + \mathrm{Re} R_1(0, \Omega_{g1}^2)]^{1/2}$; however, because the leading terms in the quantities $|m_{j_1}^{(\nu)}(\mathbf{k})|^2$ in the limit $\mathbf{a}\mathbf{k} \ll 1$ are proportional to $(\mathbf{ak})^2/\Omega_1(\mathbf{k})$ for the *db* and to $(\mathbf{ak})^4/\Omega_1(\mathbf{k})$ for the pl centers, when expression (15) is used the energy gap practically coincides with the original Ω_{g1}. Consequently, the terms that play a decisive role in the way the impurity spins affect the stability of the ground magnetic state of the crystal are those of higher order than linear in the cumulant expansion of the quantity (15) (Refs. 39, 29), which describes the indirect impurity-impurity interaction due to the exchange of virtual magnons; this interaction determines the concentration broadening of the impurity levels. In principle these terms can be calculated by means of the quantity (13), as in (15). However, as the corresponding expressions are rather cumbersome, to obtain an estimate we will use a simpler procedure based on an effective Hamiltonian for impurity spins.

As will be clear below, the concentration broadening is determined by the indirect interaction between impurities at a mean distance $\bar{r} \sim ac^{-1/2}$, while the primary contribution to this interaction is given by magnons with wave vector $k \sim \bar{r}^{-1}$; relative to these magnons, the subsystem of impurity spins is adiabatically slow if the condition $c \gg (\omega_j/J)^2$ is satisfied. Since the characteristic value of this latter quantity is about 10^{-4} to 10^{-3}, we can assume that this condition is fulfilled in the region of concentrations $c \sim 10^{-2}$, which is of primary interest to us.

Let us begin with the Hamiltonian $\mathscr{H}$, which we use to write equations of motion for the Green's functions of the form $\langle\langle \sigma_{\mathbf{p}_j}^+ | \sigma_{\mathbf{p}_j}^- \rangle\rangle_\omega$, to second order in the coupling constant $m_{j\mu}^{(\nu)}(\mathbf{k})$. It is not difficult to verify that these equations can be obtained from the following effective Hamiltonian in the adiabatic limit $\omega \to 0$:

$$\mathscr{H}_{eff} = \sum_{\mathbf{p}_j} \Big[\omega_j \sigma_{\mathbf{p}_j}^z + \sum_{\mathbf{p}'_{j'}} (V^x_{\mathbf{p}_j \mathbf{p}'_{j'}} \sigma^x_{\mathbf{p}_j} \sigma^x_{\mathbf{p}'_{j'}} + V^y_{\mathbf{p}_j \mathbf{p}'_{j'}} \sigma^y_{\mathbf{p}_j} \sigma^y_{\mathbf{p}'_{j'}}) \Big], \tag{16}$$

where the parameters of the effective anisotropic exchange interaction have the form

$$\overset{x}{V}{}^{y}_{\mathbf{p}_j \mathbf{p}_{j'}'} = \frac{2}{N} \sum_{\mathbf{k},\mu} \exp[i\mathbf{k}(\mathbf{p}_j - \mathbf{p}_{j'}')] \{ m_{j\mu}^{(1)}(\mathbf{k}) [m_{j'\mu}^{(1)}(\mathbf{k})]^* + m_{j\mu}^{(2)}(\mathbf{k}) [m_{j'\mu}^{(2)}(\mathbf{k})]^* \pm m_{j\mu}^{(1)}(\mathbf{k}) m_{j'\mu}^{(2)}(\mathbf{k}) \pm [m_{j\mu}^{(1)}(\mathbf{k})]^* [m_{j'\mu}^{(2)}(\mathbf{k})]^* \} \Omega_\mu^{-1}(\mathbf{k}) + \text{c.c.} \tag{17}$$

It is obvious that the most important contribution to the broadening of the impurity levels ω_j comes from the interaction (17) between centers of the same type:

$$\overset{x}{V}{}^{y}_{\mathbf{p}_j \mathbf{p}_j'} \equiv \overset{x}{V}{}^{y}_{j}(\mathbf{p} - \mathbf{p}') = \frac{4}{N} \sum_{\mathbf{k},\mu} \exp[i\mathbf{k}(\mathbf{p}_j - \mathbf{p}_j')] [|m_{j\mu}^{(1)}(\mathbf{k})| \pm |m_{j\mu}^{(2)}(\mathbf{k})|]^2 \Omega_\mu^{-1}(\mathbf{k}). \tag{18}$$

Calculation of these quantities, taking into account the definitions (7) and (13), shows that the dominant contribution is here determined by the following expression:

$$V_j^x(\mathbf{p} - \mathbf{p}') = \begin{cases} \dfrac{J'^2 sz}{4\pi J} \left(\dfrac{a}{|\mathbf{p} - \mathbf{p}'|} \right)^2 \sin 2\tilde{\varphi}, & \text{(19a)} \\[2ex] \dfrac{J'^2 sz}{4\pi J} \left(\dfrac{a}{|\mathbf{p} - \mathbf{p}'|} \right)^2 \cos 2\tilde{\varphi}, & \text{(19b)} \\[2ex] \dfrac{3J'^2 sz}{128\pi J} \left(\dfrac{a}{|\mathbf{p} - \mathbf{p}'|} \right)^4 \cos 4\tilde{\varphi}, & \text{(19c)} \end{cases}$$

where $\tilde{\varphi}$ is the angle between the radius vector $\mathbf{p} - \mathbf{p}'$ and the AFM vector of the $2D$ host. As for the quantity $V_j^{(y)}(\mathbf{p} - \mathbf{p}')$, it differs from $V_j^{(x)}(\mathbf{p} - \mathbf{p}')$ given in (19) by an additional small factor $\sin^2\theta_j = (\omega_j/J')^2 \ll 1$, so that the effective Hamiltonian (16) can be approximately treated as if it were an Ising Hamiltonian.

It is interesting to note that the same (Ising-like) behavior is also characteristic of the effective interaction between so-called orthogonal impurity centers in $3D$ magnets; however, this interaction, which is caused by strongly anisotropic substitutional impurities, differs from (19) by a sign change and the decay law $\propto r^{-1}$ (Ref. 29). Likewise, in insulating glasses the indirect interaction between two-level systems, which is caused by the exchange of virtual phonons, falls off as r^{-3} (Ref. 36).

5. PHASE STATES OF THE SYSTEM AT LOW TEMPERATURES

Due to the random locations of the impurity centers, an exact diagonalization for the effective Hamiltonian (16) is impossible, and we can only assert that the presence of the interaction (19) between impurity spin excitations leads to the appearance of a certain concentration broadening Δ_j of the original levels ω_j; it is important, however, to note that for the short-range interaction the correct value of this broadening cannot be obtained by using the simple mean-field approximation, that is, by self-consistent replacement of $\sigma^x_{\mathbf{p}'}$ by $\langle\sigma^x_{\mathbf{p}'}\rangle$ in (16) and calculation of the dispersion of the random quantity $\omega_{pj} = (\omega_j^2 + h_{\mathbf{p}}^2)^{1/2}$, where $h_{\mathbf{p}} = \Sigma_{\mathbf{p}'} V_j^{(x)}(\mathbf{p}-\mathbf{p}')\langle\sigma^x_{\mathbf{p}'}\rangle$. In a first-principles approach the quantity Δ_j is determined by the minimum width of the energy region around ω_j where this or some other cumulant expansion for Green's functions of the form $\ll\sigma_{\mathbf{p}}^+|\sigma_{\mathbf{p}}^-\gg$ becomes divergent. By writing the equations of motion for the latter using the Hamiltonian (16) and analyzing the behavior of the first term of the corresponding series, we can obtain conditions for convergence (at small concentrations, where $\Delta_j \ll \omega_j$) in the form[40]

$$c\left|\sum_{\mathbf{n}\neq 0} \frac{[A^{(j)}_{0\mathbf{n}}(\omega)]^2}{1-[A^{(j)}_{0\mathbf{n}}(\omega)]^2}\right| < 1, \tag{20}$$

where

$$A^{(j)}_{0\mathbf{n}}(\omega) = V_j^x(\mathbf{n})/2(\omega-\omega_j).$$

Taking into account expression (19), we obtain from (20) the following estimate for the required broadening:

$$|\omega-\omega_j| < \Delta_j = \begin{cases} c\dfrac{J'^2sz}{8J} & \text{for} \quad \text{db}, \\ 3c^2\left[\dfrac{\Gamma(^3/_4)}{4}\right]^4 \dfrac{J'^2sz}{4J} & \text{for} \quad \text{pl}, \end{cases} \tag{21}$$

i.e., in order of magnitude it equals the absolute value of the angle φ of the interaction between impurities averaged over angle at the average distance. Since the energy ω_j is the same order of magnitude for both types of centers, it is clear from (21) that the most effective concentration broadening is for levels with the dumbbell coupling. It is interesting to note that the insertion of an Me^{2+} ion into the lattice causes a shift of the "vertex" oxygen ions into the tetrahedral position; this has been confirmed by experiment.[41] In other words, a db center is always created near a pl-center. Perhaps this explains why the disruption of AFM ordering is equally rapid for both types of center.

When the concentration of impurities attains a value

$$c_1 \sim \omega_{db} J/J'^2sz \tag{22}$$

the quantity Δ_{db} becomes comparable with ω_{db} and Eq. (21) turns out to be applicable only for order-of-magnitude purposes. In this case, the concentration broadening washes out all the low-frequency part of the spectrum, including the gap in the magnon spectrum[2)] (if we keep in mind that $\omega_{db} > \Omega_{g1}$). As a result, the original ground state, for which the spins of the host form a Néel configuration in the basal plane, while the impurity spins are oriented perpendicular to this plane, no longer corresponds to the lowest energy.

For sufficiently low temperatures and a sign-varying interaction between the randomly located spins, it is well known that the energy minimum corresponds to ordering of the "spin-glass" type,[42] and in the present case this is the ordering that should characterize the impurity spins. It is significant that for $\omega_j \sim \Omega_{g1}$ and $c > c_1$, long-range order is absent not only in the impurity spins but in the host spins as well. The natural energy parameter that characterizes the low-frequency region of the spectrum in this case is the interaction energy between impurity spins at the average distances; in the present case we may choose Δ_{db} as this energy from (21). This value also determines the order of magnitude of the temperature T_f for freezing-in of the spins, which therefore increases linearly with c for $c > c_1$:

$$T_f \sim cJ'^2/J. \tag{23}$$

For $T < T_f$ band states do not exist in the magnon spectrum with wave vectors smaller than the minimum value

$$k_{min} \sim ca^{-1}(J'/J)^2, \tag{24}$$

while the value of the quantity inverse to this,

$$\xi_{2D} = 2\pi k_{min}^{-1} \sim ac^{-1} \gg a \tag{25}$$

determines the low-temperature correlation length in the $2D$ subsystem of host spins.

We note further that if the parameters of the system were such that the inequality $\omega_j \ll \Omega_{g1}$ held (e.g., in the absence of the Dzyaloshinskii interaction), then its ground state should be characterized by spin-glass ordering of the impurity spins superposed on the original long-range order for the host spins. This situation, in particular, occurs for AFM with quadrupole impurities.[28,29]

It follows from the estimates (22), (23), and will be confirmed below by other estimates, that the region in which the spin-glass type of ordering exists is bounded from above by a temperature on the order of 10 K, i.e., far below T_N. These estimates are in satisfactory agreement with the experimental estimates from Ref. 43 as well.

For $T > T_f$, the impurity-host interaction is "switched off" due to thermal saturation of the two-level impurity systems (i.e., $\sigma \to 0$); the subsystem of impurity spins becomes paramagnetic, and, as it turns out, does not affect the AFM of the host to any appreciable degree. As a result of this, the long-range order of the host must be reestablished, i.e., a reentrant phase transition takes place. The change in T_N due to the interaction between local spin excitations is only of order cT_N.

Thus, within the framework of the system (4) and (12), we find that disruption of AFM order in lightly-doped La_2CuO_4 is possible only in the range of very low temperatures. Then in order to resolve the question of whether or not this disruption is possible in the temperature interval $T_f < T < T_N$, we must investigate some other mechanisms by which impurities can affect the ground magnetic state of the crystal.

6. STATIC DISTORTIONS OF THE LATTICE AND THE DISRUPTION OF LONG-RANGE MAGNETIC ORDER

In the final analysis, the inability of the mechanism discussed in the previous section to influence the AFM order in a crystal for $T \gg T_f$ is related to the smallness of the eigenfrequencies with which impurity spins precess; therefore, in order to explain the observed behavior of the magnetic subsystem, we must seek some other way to disrupt the long-range order, i.e., due to those impurity perturbations that "survive" even at high temperatures. Perhaps the most obvious source of such perturbations is the static strains in the crystal lattice that arise when a lanthanum ion is replaced by an alkali-earth element, due to the difference in the corresponding ionic radii, or when above-stoichiometry oxygen is injected into the lattice. These strains do not depend on the spin state of the impurity, and their coupling to the host spins arises from their strong local effect on the spin anisotropy of the host. In this context it is relevant to recall (see Sec. 3 above) the original weak anisotropy ΔJ_{rh} (denoted from here on by ΔJ for simplicity), which determines the stability of the ground state of the $2D$ AFM, is completely due to the weak uniform "strain" φ of the perovskite structure in La_2CuO_4. In this case, the appearance of a local distortion φ_j near an impurity center of jth type, which differs strongly from the uniform strain both in magnitude $\varphi_j \gg \varphi$ and in symmetry, can lead to a very significant perturbation of the center.

A detailed microscopic analysis of these processes[44] shows that a real possibility exists for strong local changes in the parameters of the antisymmetric exchange interaction; however, the structure which results from these perturbations is found to be too complicated for direct use, e.g., in calculating Green's functions. Nevertheless, the necessary qualitative conclusions can be obtained even in the limit of the simplest phenomenological approach, which assumes the presence in the crystal of centers that are random with regard to direction of the intraplanar anisotropy, which is characterized by the quantity $\delta J \gg \Delta J$.

In order to investigate the large-scale spin fluctuations, we describe the spin configuration of the host in the continuum approximation, using the angle $\psi(\mathbf{r})$ which the AFM vector makes at the point $\mathbf{r}$ with the direction of the intraplanar anisotropy of the host, and in accordance with Hamiltonian (3) and the assumptions we have made regarding the character of the impurity-induced effects, we use the following phenomenological spin energy density:

$$\mathscr{E}(\psi(\mathbf{r}))$$

$$=\frac{1}{2}J[\nabla\psi(\mathbf{r})]^2+[\sin\psi(\mathbf{r})]^2\left[\frac{\Delta J}{a^2}+\delta J\sum_{\mathbf{p}}\sigma_{\mathbf{p}}\delta(\mathbf{r}-\mathbf{p})\right], \tag{26}$$

where $\mathbf{p}$ runs over the random positions of the impurity centers and $\sigma_{\mathbf{p}}$ is a random quantity which takes on the values ± 1 with equal probability.

It is apparent from an analysis of the first two terms on the right side of (26) that the characteristic scale of ρ for fluctuations $\psi_\rho(\mathbf{r})$ is the quantity $r_B = a\sqrt{J/\Delta J}$, i.e., the Bloch length. If the impurity concentration satisfies the adiabatic condition mentioned in Sec. 4, i.e., $c \gg (\omega_j/J)^2$, then we have $r_B \gg \bar{r}$, and on the average a large number of impurities are found within a single fluctuation. By considering the energy $E(\boldsymbol{\rho}) = \int \mathscr{E}(\psi_\rho(\mathbf{r}))d\mathbf{r} = \alpha_1 J + \alpha_2 \Delta J(\rho/a)^2 + S(\boldsymbol{\rho})$ associated with such a fluctuation, and using for the random quantity $S(\boldsymbol{\rho}) = (\delta J/a)\int\Sigma_{\mathbf{p}}\sigma_{\mathbf{p}}\delta(\mathbf{r}-\mathbf{p})\sin^2\psi_\rho(\mathbf{r})d\mathbf{r}$ the obvious Gaussian distribution

$$P_{\rho}(S)=\exp\left[-\frac{S^2}{\alpha_3 c(\rho\delta J/a)^2}\right]\Big/\ (\pi\alpha_3 c)^{1/2}\frac{\rho\delta J}{a},$$

[where the constants $\alpha_{1,2,3} \sim 1$ are in general determined by optimizing the trial function $\psi_\rho(\mathbf{r})$], we can determine the probability $W(T,c)$ of fluctuation-induced disruption of the original spin order at an arbitrary point in the crystal for a given temperature and concentration. Then the function $T_N(c)$ is found from the condition $W(T_N(c),c) = \eta$, where $\eta \sim 1/2$. This function is found to be simplest in the limiting cases of small concentrations of low temperatures.

At small concentrations (but nevertheless with $c \gg (\omega_j/J)^2$), when the width of the distribution $P_\rho(S)$ is small compared to $\alpha_1 J + \alpha_2 \Delta J(\rho/a)^2$, a special role in the disruption of long-range order is played by temperature-induced fluctuations, which in this case can be treated as statistically independent of the concentration-induced fluctuations, and consequently regarded as independent among themselves at various points in the crystal. The corresponding contribution to $W(T,c)$ can be found by summing the probabilities (equal among themselves) for all possible thermal fluctuations of radius $|\boldsymbol{\rho}|$ trapped at a given point (leading finally to multiplication of the result by $(\rho/a)^2$), and subsequent optimization with respect to the radius of the fluctuation:

$$W(T,c)=\max_{|\boldsymbol{\rho}|}\int_0^\infty dE(\rho/a)^2 e^{-E/T}g(E,\boldsymbol{\rho}), \tag{27}$$

where

$$g(E,\ \boldsymbol{\rho})=P_{\rho}[\alpha_1 J+\alpha_2\Delta J(\rho/a)^2-E]. \tag{28}$$

The maximum in (27) corresponds to $\rho_{\max} \approx aT[\alpha_2\Delta J(T-\alpha_1 cJ/c_2)]^{-1/2}$, and the corresponding function $T_N(c)$ turns out to be linear:

$$T_N(c)\approx T_N(0)(1-c/c_2), \tag{29}$$

where $T_N(0)$ corresponds to the value of T_N determined in (8), while

$$c_2=4\alpha_1\alpha_2 J\Delta J[\alpha_3(\delta J)^2]^{-1}$$

is a characteristic concentration at which (as we will show below) long-range magnetic order is disrupted in the system. We note that the quantities c_1 of (22) and c_2 are in general independent of each other, and it is necessary for AFM and spin-glass order to be able to coexist in some range of concentrations that the inequality $c_1 < c_2$ hold, i.e.,

$$\omega_{db}<2\alpha_1\alpha_2 J'^2\Omega_{g1}{}^2[\alpha_3(\delta J)^2 Jsz]^{-1}.$$

As the concentration increases, the quantity $\rho_{\max}$ increases as well, eventually attaining a value of r_B, while the transition temperature decreases. As $c \to c_2$ and $T \to 0$, a decisive role is played by statistical concentration-induced fluctuations, for which the probabilities of disrupting the original order within a single fluctuation cluster are rigorously correlated; this should lead to elimination of the factor $\boldsymbol{\rho}^2$. For $T = 0$, the quantity $W(0,c)$ is determined simply by the total probability for forming "flipped" ($\psi = \pi/2$) clusters corresponding to negative E:

$$W(0,c)=\max_{|\rho|}\int_{-\infty}^{0} dE\, g(E,\rho)=\frac{1}{2}\left[1-\Phi\left(\left(\frac{c_2}{c}\right)^{1/2}\right)\right], \quad (30)$$

where

$$\Phi(x)=\frac{2}{\pi^{1/2}}\int_0^x \exp(-t^2)\,dt$$

is the probability integral. From (30) it follows that $T_N(c)$ vanishes, i.e., long-range order in the system completely collapses, in practice at concentrations on the order of c_2. In fact, the decrease in T_N is bounded at a point c_2' close to c_2, at which $T_N(c)$ reaches the value $T_f(c)$ given in (23).

As $T\to 0$, the positions of the boundaries of the region of long range order found above from our phenomenological study are found to agree with the results of the following model microscopic approach. In fact, by using in place of (12) a Hamiltonian for perturbations of the anisotropy due to induced static strains at impurity centers, e.g., of pl-type,$^{3)}$ along with the original magnon Hamiltonian (4),

$$\mathscr{H}_{an}=-\delta J\sum_{\mathbf{p},\alpha}\{[S^{x}_{(\mathbf{p}+\mathbf{a}/2)_\alpha}]^2+[S^{x}_{(\mathbf{p}-\mathbf{a}/2)_\alpha}]^2+[S^{z}_{(\mathbf{p}+\mathbf{b}/2)_\beta}]^2$$

$$+[S^{z}_{(\mathbf{p}-\mathbf{b}/2)_\beta}]^2\},\quad \beta\neq\alpha, \quad (31)$$

and taking into account only magnon excitations from the lower branch of the magnon spectrum with $\mu = 1$, we can reduce the resulting Hamiltonian to a magnon-scattering Hamiltonian of the form

$$\mathscr{H}_{an}=\frac{1}{N}\sum_{\mathbf{k},\mathbf{k}',\mathbf{p}}\exp[i(\mathbf{k}-\mathbf{k}')\mathbf{p}]\,\{V(\mathbf{k},\mathbf{k}')\beta_1^+(\mathbf{k})\beta_1(\mathbf{k}')$$

$$+{}^1/_2 W(\mathbf{k},\mathbf{k}')[\beta_1(\mathbf{k})\beta_1(\mathbf{k}')+\beta_1^+(\mathbf{k})\beta_1^+(\mathbf{k}')]\}, \quad (32)$$

where the scattering parameters equal:

$$V(\mathbf{k},\mathbf{k}')=\delta J\left[(u_{1\mathbf{k}}+v_{1\mathbf{k}})(u_{1\mathbf{k}'}+v_{1\mathbf{k}'})\cos\frac{(\mathbf{k}-\mathbf{k}')\mathbf{a}}{2}\right.$$

$$\left.-2(u_{1\mathbf{k}}u_{1\mathbf{k}'}+v_{1\mathbf{k}}v_{1\mathbf{k}'})\cos\frac{(\mathbf{k}-\mathbf{k}')\mathbf{b}}{2}\right],$$

$$W(\mathbf{k},\mathbf{k}')=\delta J\left[(u_{1\mathbf{k}}+v_{1\mathbf{k}})(u_{1\mathbf{k}'}+v_{1\mathbf{k}'})\cos\frac{(\mathbf{k}-\mathbf{k}')\mathbf{a}}{2}\right.$$

$$\left.-2(u_{1\mathbf{k}}v_{1\mathbf{k}'}+u_{1\mathbf{k}'}v_{1\mathbf{k}})\cos\frac{(\mathbf{k}-\mathbf{k}')\mathbf{b}}{2}\right].$$

As $T\to 0$ it is not difficult to obtain the following dispersion equation from (32):

$$\omega^2-\Omega_1^2(\mathbf{k})-R_{\mathbf{k}}(\omega)=0, \quad (33)$$

where

$$R_{\mathbf{k}}(\omega)=\frac{2c\Omega_1(\mathbf{k})}{N}\sum_{\mathbf{k}'}\frac{\Omega_1(\mathbf{k}')[|V(\mathbf{k},\mathbf{k}')|^2+|W(\mathbf{k},\mathbf{k}')|^2]}{\omega^2-\Omega_1^2(\mathbf{k}')}$$

equals

$$R_0(0)=cR_{def}(\delta J)^2,$$

$$R_{def}=\left(\frac{4}{\pi}\right)^2\int_0^\pi\int_0^\pi dx\,dy\,\frac{[\cos(x/2)-\cos(y/2)]^2}{1-\cos(x/2)\cos(y/2)}\approx 6{,}04$$

for $\mathbf{k}=0$ and $\omega\to 0$. An analogous result is obtained for the db-centers as well.

It is easy to see from (33) that, by virtue of the definition (7), the gap in the magnon spectrum reduces to zero, i.e., long-range order in the system is disrupted, when

$$c\to c_{2m}=2J\Delta J(sz)^2/(\delta J)^2R_{def},$$

which is in fair agreement with the quantity c_2 determined above (see Ref. 29), and indicates agreement between the phenomenological and model microscopic approaches.

We note in this case that the continuous function $T_N(c)$, which agrees qualitatively with the results of the phenomenological description, can be obtained by substituting into Eq. (8) the concentration-dependent value of the gap in the crystal with impurities determined from Eq. (33):

$$\Omega_{g1}(c)=\Omega_{g1}(1-c/c_{2m})^{1/2}.$$

This implies the following result [compare (8)]:[25]

$$T_N(c)=T_N(0)\,\frac{1-{}^1/_2\ln(1-c/c_{2m})}{\ln(J/D)}.$$

As the impurity concentration increases further, so that we have $c>c_2$, there is no long-range order in the system at any temperature; however, in the low-temperature region ($T<T_f$) the effective impurity-impurity interaction (17) still remains, since the primary contribution to this interaction comes from magnons with rather well-defined wave vectors: $k\sim\bar{r}^{-1}\sim\sqrt{c}/a\gg k_{\min}\sim\sqrt{c\delta J}/aJ$. However, in this case the presence in the host of a large number of "flipped" fluctuation clusters, which are simultaneously subject to strong temperature-induced fluctuations, can result in loss of the equilibrium state for impurity spins, and as a consequence a bound on the increase of the function $T_f(c)$ for $c>c_2$ [relative to (23)].

The overall shape of the phase diagram for this system in "temperature-concentration" variables is shown in Fig. 3. We note that the diagram obtained in this way is in complete agreement with the diagrams given in Refs. 1, 43, and 46, which are constructed by analyzing the various experimental data.

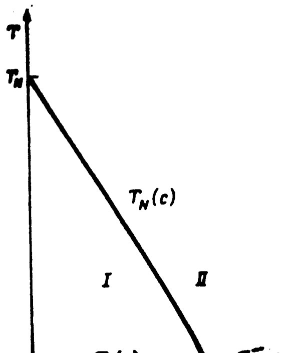

FIG. 3. Qualitative form of the magnetic phase diagram of lightly-doped La_2CuO_4. I, II are regions of the AFM and paramagnetic phases, while the dashed region is for a spin-glass type phase.

7. CONCLUSIONS

The analysis we have carried out in this paper shows that the $La_{2-x}Me_xCuO_4$ system (like La_2CuO_{4+y}) possesses diverse magnetic properties that are characterized by alternation of AFM order, spin-glass order, and paramagnetic states. In this case, in the various regions of the spectrum there exist both spin excitations of band type (magnons) and impurity fluctuation-induced states. An additional important feature of this system is the presence in it of strongly-developed AFM correlations over rather long distances $\xi_{2D} \ll k_{\min}^{-1}$, i.e., large AFM clusters, even in the paramagnetic phase. However, the quasi-$2D$ character of the ordering can lead to activation energies for rotating the total moment of such a cluster that turn out to be independent of the cluster size and very much smaller than in a system with $3D$ order. Therefore, long-time correlations in these clusters can perhaps be observed experimentally at very low temperatures.

According to our calculations, the existence of localized spins in the system cannot disrupt the long-range AFM order in an irreversible fashion, and the field of random strains caused by the dopants turns out to be more effective in doing this. In principle, this mechanism for suppression can operate in other HTSC as well, in particular in ceramic 123. However, in this material centers of an entirely different type appear: each insertion of an oxygen ion into $YBa_2Cu_3O_{6+x}$ gives rise to an exchange pair of strongly bound Cu^{2+} spins (i.e., a dumbbell) in the Cu–O chains, which in turn not only frustrates the AFM ordering in the vicinity of the CuO_2 planes due to the rather weak interplanar exchange interaction, but also gives rise to a local rhombic distortion of the lattice.

In this connection, however, we note finally that it is more likely that such a "strain-induced" disappearance of magnetic order will occur in the α-phase of solid oxygen, which is a quasi-$2D$ AFM solid in which the addition of a relatively small number of N_2 (or Ar) impurities completely destroys the long-range spin $3D$ order.[47]

We are grateful to S. V. Maleev, S. G. Ovchinnikov, S. M. Ryabchenko, and S. K. Tolpygo for useful discussions of various aspects of the problem of disruption of long-range magnetic order in HTSC.

[1)] This does not apply to "n-type" HTSC, where the destruction of magnetism is most likely described by the models investigated by Wiegmann,[17] although it is perhaps necessary to include in these models the local character of the states which arise as a result of doping as well.

[2)] It is also not difficult to verify that at this concentration there is a formal sign change of the coefficient of $(\mathbf{ak})^2$ in the dispersion law for spin waves, as follows from (14). In other words, due to the renormalization the spin wave velocity becomes negative. However, since the states in this region are in fact localized, we cannot even speak of a dispersion law in general.

[3)] Here, for simplicity we have written the anisotropy of the spins near an impurity atom caused by the rotation of the octahedra in single-ion form, which strictly speaking has meaning only for $S \geqslant 1$. In actuality, however, for the system under discussion here, where we have $S \neq 1/2$, the spin anisotropy is caused by the anisotropic exchange interaction. Nevertheless, the results obtained here also remain valid for this type of anisotropy (if we treat δJ as the anisotropic exchange interaction constant). This is associated with the fact that for $S \neq 1/2$ the quasiclassical approach used here, which is correct in the case of a single-ion anisotropy that is weak compared to the exchange interaction, gives results equivalent to the case of inter-ion anisotropy or the antisymmetric exchange interaction.

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