Quantum fluctuations in the frustrated antiferromagnet $Sr_2Cu_3O_4Cl_2$

A. B. Harris
Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, Pennsylvania 19104

A. Aharony, O. Entin-Wohlman, and I. Ya. Korenblit
School of Physics and Astronomy, Raymond and Beverly Sackler Faculty of Exact Sciences, Tel Aviv University, Tel Aviv 69978, Israel

R. J. Birgeneau
Center for Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

Y.-J. Kim
Division of Engineering and Applied Science, Harvard University, Cambridge, Massachusetts 02139
and Center for Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 20 September 2000; published 22 June 2001)

$Sr_2Cu_3O_4Cl_2$ is an antiferromagnet consisting of weakly coupled CuO planes which comprise two weakly interacting antiferromagnetic subsystems I and II which order at respective temperatures $T_{\rm I}\approx 390$ K and $T_{\rm II}\approx 40$ K. Except asymptotically near the ordering temperature, these systems are good representations of the two-dimensional quantum spin-1/2 Heisenberg model. For $T<T_{\rm II}$ there are four low-energy modes at zero wave vector, three of whose energies are dominated by quantum fluctuations. For $T_{\rm II}<T<T_{\rm I}$ there are two low-energy modes. The mode with lower energy is dominated by quantum fluctuations. Our calculations of the energies of these modes (including dispersion for wave vectors perpendicular to the CuO planes) agree extremely well with the experimental results of inelastic neutron scattering (in the accompanying paper) and for modes in the sub-meV range observed by electron spin resonance. The parameters needed to describe quantum fluctuations are either calculated here or are taken from the literature. These results show that we have a reasonable qualitative understanding of the band structure of the lamellar cuprates needed to calculate the anisotropic exchange constants used here.

DOI: 10.1103/PhysRevB.64.024436 PACS number(s): 76.50.+g, 75.10.Jm, 75.50.Ee

I. INTRODUCTION

There has been a resurgence of interest in low-dimensional magnetism due in part to the desire to understand high-T_c superconductivity. The lamellar copper oxide systems, when suitably doped give rise to a family of superconductors with T_c's in the range about 30 K.[1] In these systems the Cu ions are essentially in a $3d^9$ configuration. Due to a large on-site Coulomb interaction, the states of this system which are accessible at ambient temperature have one hole per Cu ion, and hence the manifold of such accessible states is described by a spin-1/2 Hamiltonian having antiferromagnetic interactions, which are strongest between nearest-neighboring Cu ions in the CuO_2 plane. That this system is a nearly perfect realization of the two-dimensional (2D) spin 1/2 quantum Heisenberg model has been established by a wide variety of experiments.[2]

Recently, a variant of this system $Sr_2Cu_3O_4Cl_2$ (2342) has been shown to display very interesting magnetic properties.[3–5] The structure of this system[6] is one in which an additional Cu ion (which we refer to as a $Cu_{\rm II}$ ion) is inserted at the center of alternate Cu plaquettes of the usual copper lattice, whose ions we refer to as $Cu_{\rm I}$'s. Although all the Cu ions are chemically equivalent, they play very different roles insofar as magnetism is concerned. The $Cu_{\rm I}$'s order at a relatively high temperature ($T_I=386$ K) and have properties similar to those of other lamellar cuprate antiferromagnets.[2] With respect to the isotropic exchange interactions, the coupling between $Cu_{\rm I}$ and $Cu_{\rm II}$ ions is frustrated. As a result, the $Cu_{\rm II}$'s order independently at a much lower temperature, $T_{\rm II}=39.6$ K into the magnetic structure shown in Fig. 1. For $T_{\rm II}<T<T_{\rm I}$ a very small residual anisotropic exchange interaction causes the $Cu_{\rm II}$ spins to have a small ferromagnetic moment, the study of which[4] led to the determination of the magnetic structure which has recently been confirmed by neutron diffraction.[7] The study of the statics also led to the determination of several coupling constants in the Hamiltonian used to model this system.

A natural continuation of this study was to investigate the dynamics of this system, and in the accompanying paper[7] (which we refer to as paper I) an inelastic neutron scattering study of this system is reported. One interesting result of these experiments was that although the coupling between the $Cu_{\rm I}$'s and $Cu_{\rm II}$'s is frustrated in the mean-field sense, the spin-wave spectrum showed an incontrovertible signature of interactions between these subsystems.[5,7] The nature of this coupling was described by Shender in a seminal paper.[8] Although this phenomenon has been identified in other materials,[9] the effect of this coupling, caused by quantum fluctuations, is perhaps the most dramatic in the system 2342, as described briefly previously[5] and in more detail in paper I. As the $Cu_{\rm II}$ system orders for $T<T_{\rm II}$, the small gap spin-wave energies are found to increase sharply. This increase indicates that even though the $Cu_{\rm I}$-$Cu_{\rm II}$ coupling is frustrated in the mean-field sense, quantum fluctuations lead

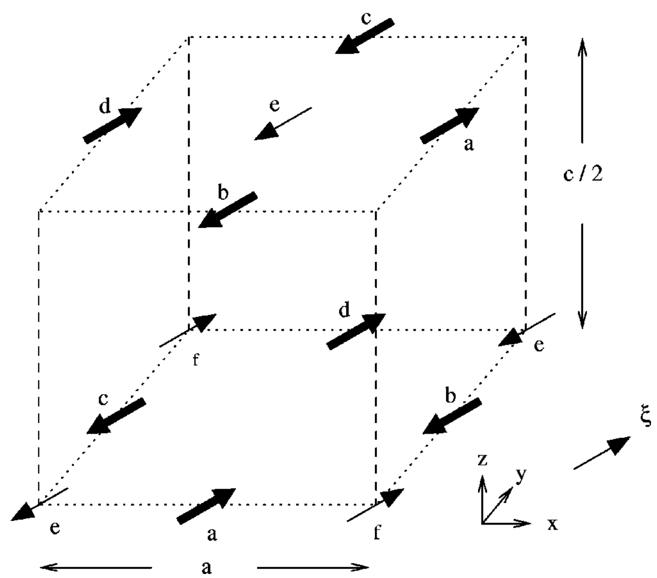

FIG. 1. Magnetic structure of 2342. The Cu_I spins (in sublattices $\underline{a}$, $\underline{b}$, $\underline{c}$, and $\underline{d}$) are thick arrows and the Cu_{II} spins (in sublattices $\underline{e}$ and f) are thin arrows. The basis vectors for the magnetic unit cell are $\mathbf{a}_1 = a(\hat{x}+\hat{y})$, $\mathbf{a}_2 = a(\hat{x}-\hat{y})$, and $\mathbf{a}_3 = \frac{1}{2}(a\hat{x}+a\hat{y}+c\hat{z})$. All spin directions are in the CuO (x-y) plane. The ξ axis is defined to be collinear with the spin directions.

to a significant interaction between sublattices. A less obvious type of frustration arises with respect to the in-plane anisotropy associated with the bond anisotropy of the exchange interactions. When the moments lies in the easy plane, the exchange tensor for spins i and j in the plane has different values for directions parallel and perpendicular to the i-j bond. However, within mean field theory this anisotropy disappears when the average over all bonds is taken. But as before, there is a significant residual interaction due to quantum fluctuations which gives rise to in-plane anisotropy. Finally, even classically frustration can be removed by exchange anisotropy which has a form similar to the dipolar interaction. We will refer to such exchange anisotropy as pseudodipolar.

The purpose of the present paper is to calculate the spin-wave spectrum in order to give a theoretical interpretation to the data presented in paper I. From the discussion so far it is clear that most of these phenomena are outside the scope of linearized spin-wave theory. What is required is a nonlinear spin-wave analysis, i.e., an analysis which includes the effects of quantum fluctuations. In fact, from an analysis of the magnetic structure of the cuprates[10] it was shown that there are several perturbations away from the linear analysis of the isotropic Heisenberg model that one must consider. These are the ones mentioned above, namely, (a) quantum fluctuations of otherwise frustrated interactions, (b) quantum fluctuations of the anisotropic in-plane exchange interactions, and (c) pseudodipolar exchange anisotropy between the Cu_I and Cu_{II} subsystems. In a simplified way, one can categorize these effects in the way they contribute to the spin-wave energies, which is given by the famous formula[11]

$$\omega = \sqrt{2H_E H_A}, \tag{1}$$

where H_E (H_A) is the exchange (anisotropy) field and we work in units such that ω, H_E, and H_A are all energies, usually given in meV (1 meV/k_B=11.6 K, 1 meV/h =241.8 Ghz.) We will see that the out-of-plane anisotropy of the exchange interactions gives rise to a corresponding out-of-plane anisotropy field H_A^{out} which has been understood in terms of the out-of-plane anisotropy in the exchange interactions without reference to fluctuations.[12,13] In contrast, the in-plane anisotropy of the exchange interactions, when summed over bonds, averages to zero and therefore only contributes when fluctuations are taken into account.[12,13] The mechanism studied by Shender[8] contributes to H_A except for the Goldstone mode, whose energy becomes nonzero only when lattice anisotropy is introduced.

One might expect that the number of coupling constants might be so large that no useful information or test of the theory would be possible. As it happens, the fit to the energy of the gaps is overdetermined and the agreement between theory and experiment in some instances is quite remarkable, as can be seen in paper I. The observation of the modes whose energy depends on the in-plane anisotropy leads to the determination of the in-plane anisotropy of the exchange interactions. These quantities are difficult to obtain experimentally. Their values can be compared to calculations[12–14] based on the electronic structure of the cuprates the knowledge of which may lead to a better understanding of the high-T_c superconductors.

One should recognize that at the moment inelastic neutron scattering does not easily detect modes in the sub-meV range of energy. As a result neutron scattering experiments have not detected those in-plane modes whose energy depends only on the in-plane anisotropy. Recently, however, the modes in the sub-meV range of energy have been observed by ESR experiments of the group at RIKEN.[15,16] The mere existence of these modes tends to confirm the spin-wave calculations. Moreover, the fact that they are found in the predicted range of energy strongly supports the theoretical calculations in this paper.

Briefly, this paper is organized as follows. In Sec. II the Hamiltonian with its various anisotropic exchange interactions is specified. In Sec. III we start by discussing briefly the framework within which the calculations are to be done and we give the Dyson-Maleev transformation[17] to boson operators. In Sec. IV the isotropic exchange Hamiltonian is discussed, first within harmonic theory and then including spin-wave interactions, which are essential to obtain a qualitatively correct spectrum. In Sec. V the various anisotropies are included in an effective quadratic spin-wave Hamiltonian. In Sec. VI we give explicit results for the spin-wave energies for the case when the transverse wave vector is zero and show the comparison of our calculations with the recent experiments of the MIT group. In Sec. VII intensities of modes are discussed, with numerical results given for zero wave vector relative to the Bragg peaks for Cu_I and Cu_{II}. Our conlusions are summarized in Sec. VIII.

II. HAMILTONIAN

The Hamiltonian that we intend to treat is written as

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2, \tag{2}$$

where $\mathcal{H}_1$ includes almost all the significant interactions, namely, all the intraplanar interactions and the unfrustrated interactions between nearest neighbors in adjacent CuO planes and $\mathcal{H}_2$ includes small residual anisotropic interplanar interactions involving Cu_{II} spins. Since this latter term is totally negligible *except* for extremely small wave vector and for the lowest-energy mode, it is only necessary to include contributions from $\mathcal{H}_2$ evaluated at zero wave vector. Since the effects of $\mathcal{H}_2$ are only relevant to the extremely low frequency spectrum, we defer consideration of $\mathcal{H}_2$ until Secs. V B 4 and V B 5.

Thus we write $\mathcal{H}_1$ in tensor notation as

$$\mathcal{H}_1=\tfrac{1}{2}\sum_{\langle i,j\in I\rangle}\mathbf{S}_i\mathbf{J}_I\mathbf{S}_j+\sum_{\langle i\in I,j\in II\rangle}\mathbf{S}_i\mathbf{J}_{\text{I-II}}\mathbf{S}_j+\sum_{\langle i,j\in II\rangle}\mathbf{S}_i\mathbf{J}_{\text{II}}\mathbf{S}_j+\sum_{i\in I}J_3\mathbf{S}_i\cdot\mathbf{S}_{i+(1/2)c\hat{z}}, \tag{3}$$

where $i\in I(i\in II)$ means that site i runs over Cu_I (Cu_{II}) sites and $\langle\ \rangle$ restricts the summation to nearest neighbors of the indicated type in the same Cu-O plane. The only unfrustrated coupling between planes is that (J_3) between Cu_I's directly above or below one another. We will allow the couplings $\mathbf{J}_I$, $\mathbf{J}_{\text{I-II}}$, and $\mathbf{J}_{\text{II}}$ to be anisotropic, whereas for simplicity we take $\mathbf{J}_3$ to be isotropic. Here and below we use a hybrid notation for site labels in which the label $i+\mathbf{r}$ indicates a site at position $\mathbf{r}$ with respect to site i. In $\mathcal{H}_2$ we include the interplanar Cu_I-Cu_{II} and Cu_{II}-Cu_{II} couplings whose isotropic parts are frustrated.

We first discuss the principal axes of the exchange tensor $\mathbf{J}_I$ associated with a bond between nearest-neighboring Cu_I spins in a CuO plane. This bond is invariant with respect to two mirror planes: one in the CuO plane and the other perpendicularly bisecting the Cu_I-Cu_I bond in question. Accordingly, the principal axes of the Cu_I-Cu_I exchange tensor between nearest neighbors lie along the three crystal (1,0,0) directions, just as they would be in the absence of the Cu_{II}'s. In that case, the exchange tensor will have different values corresponding to the directions (i) along the bond in question, (ii) perpendicular to the bond in question but in the CuO plane, and (iii) along the crystal $\underline{c}$ direction. The principal axes of the other in-plane interactions are similarly fixed by symmetry.[13,19] Then the Hamiltonian $\mathcal{H}_1$ may be written as follows:

$$\begin{aligned}\mathcal{H}_1=&\tfrac{1}{2}\sum_{i\in I}\sum_{\delta_1}(J_I^zS_i^zS_{i+\delta_1}^z+J_I^{\|}[\mathbf{S}_i\cdot\hat{\delta}_1][\mathbf{S}_{i+\delta_1}\cdot\hat{\delta}_1]\\&+J_I^{\perp}[\mathbf{S}_i\cdot\hat{e}_1][\mathbf{S}_{i+\delta_1}\cdot\hat{e}_1])+\sum_{i\in II}\sum_{\delta_{2,1}}(J_{\text{I-II}}^zS_i^zS_{i+\delta_{2,1}}^z\\&+J_{\text{I-II}}^{\|}[\mathbf{S}_i\cdot\hat{\delta}_{2,1}][\mathbf{S}_{i+\delta_{2,1}}\cdot\hat{\delta}_{2,1}]+J_{\text{I-II}}^{\perp}[\mathbf{S}_i\cdot\hat{e}_{2,1}]\\&\times[\mathbf{S}_{i+\delta_{2,1}}\cdot\hat{e}_{2,1}])+\tfrac{1}{2}\sum_{i\in II}\sum_{\delta_2}(J_{\text{II}}^zS_i^zS_{i+\delta_2}^z+J_{\text{II}}^{\|}[\mathbf{S}_i\cdot\hat{\delta}_2]\\&\times[\mathbf{S}_{i+\delta_2}\cdot\hat{\delta}_2]+J_{\text{II}}^{\perp}[\mathbf{S}_i\cdot\hat{e}_2][\mathbf{S}_{i+\delta_2}\cdot\hat{e}_2])\\&+J_3\sum_{i\in I}\mathbf{S}_i\cdot\mathbf{S}_{i+\frac{1}{2}c\hat{z}},\end{aligned} \tag{4}$$

where δ_1 (δ_2) labels the nearest neighbor vectors in the plane connecting adjacent Cu_I's (Cu_{II}'s) and $\delta_{1,2}$ labels vectors in the CuO plane which give the displacements of nearest-neighboring Cu_I's relative to a Cu_{II}, and hat indicates a unit vector. Also $\hat{e}_1$, $\hat{e}_{2,1}$, and $\hat{e}_2$ are unit vectors in the CuO plane which are perpendicular to, respectively, δ_1, $\delta_{2,1}$, and δ_2.

We separate the Hamiltonian $\mathcal{H}_1$ into an isotropic part $\mathcal{H}_0$ and an anisotropic perturbation $\mathcal{H}'$. For that purpose we write

$$\Delta J_1=\tfrac{1}{2}(J_{\text{I}}^{\|}+J_{\text{I}}^{\perp})-J_{\text{I}}^z,\quad \Delta J_{12}=\tfrac{1}{2}(J_{\text{I-II}}^{\|}+J_{\text{I-II}}^{\perp})-J_{\text{I-II}}^z,$$

$$\Delta J_2=\tfrac{1}{2}(J_{\text{II}}^{\|}+J_{\text{II}}^{\perp})-J_{\text{II}}^z, \tag{5}$$

$$\delta J_1=\tfrac{1}{2}(J_I^{\|}-J_I^{\perp}),\quad \delta J_{12}=\tfrac{1}{2}(J_{\text{I-II}}^{\|}-J_{\text{I-II}}^{\perp}),\quad \delta J_2=\tfrac{1}{2}(J_{\text{II}}^{\|}-J_{\text{II}}^{\perp}), \tag{6}$$

$$\tilde{J}=\tfrac{1}{3}(J_{\text{I}}^{\|}+J_{\text{I}}^{\perp}+J_{\text{I}}^z),\quad \tilde{J}_{12}=\tfrac{1}{3}(J_{\text{I-II}}^{\|}+J_{\text{I-II}}^{\perp}+J_{\text{I-II}}^z),$$

$$\tilde{J}_2=\tfrac{1}{3}(J_{\text{II}}^{\|}+J_{\text{II}}^{\perp}+J_{\text{II}}^z). \tag{7}$$

Thus the ΔJ's describe the out-of-plane anisotropy (i.e., the energy which gives rise to an easy plane) which is responsible for the 5 meV anisotropy gap in the spin-wave spectra of cuprates which do not have Cu_{II}'s. Similarly, the δJ's describe the in-plane anisotropy (i.e., the anisotropy within the easy plane) and they (i) are responsible for the weak ferromagnetic moment[3,4] induced in the Cu_{II} subsystem by the staggered moment in the Cu_I subsystem and (ii) contribute to the macroscopic or phenomenological fourfold anisotropy constant K_4.[13,3,4] (We shall see later that $\mathcal{H}_2$ also contributes to K_4.) Note that δJ_{12} is what was called J_{pd} in Refs. 3 and 4, but differs by a factor of 2 from its definition in Refs. 13 and 14. The largest coupling is J ($J_2/J\approx J_{12}/J\approx 0.1$ and $J_3/J\approx 10^{-3}$), while the relative anisotropies $\Delta J/J$ and $\delta J/J$ are at most 10^{-3}.[13,14,3,4]

With these notations the isotropic Hamiltonian is

$$\mathcal{H}_0=\tfrac{1}{2}\sum_{i\in I}\sum_{\delta_1}\tilde{J}\mathbf{S}_i\cdot\mathbf{S}_{i+\delta_1}+\sum_{i\in II}\sum_{\delta_{2,1}}\tilde{J}_{12}\mathbf{S}_i\cdot\mathbf{S}_{i+\delta_{2,1}}+\tfrac{1}{2}\sum_{i\in II}\sum_{\delta_2}\tilde{J}_2\mathbf{S}_i\cdot\mathbf{S}_{i+\delta_2}+\sum_{i\in I}J_3\mathbf{S}_i\cdot\mathbf{S}_{i+(1/2)c\hat{z}} \tag{8}$$

and the anisotropic perturbation is

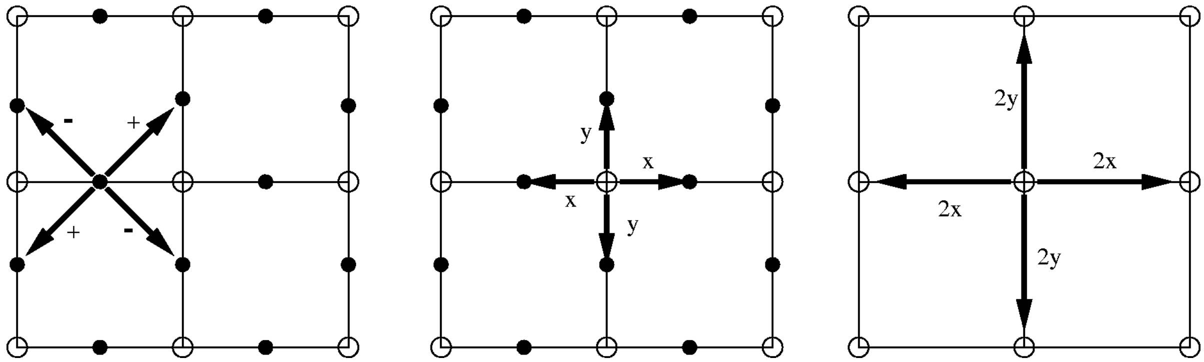

FIG. 2. Nearest-neighbor vectors connecting magnetic ions in a CuO plane. Cu_I spins are filled circles and Cu_{II} spins are open circles. Left: the vectors δ_+ and δ_- between nearest-neighboring Cu_I spins. Center: the vectors δ_x and δ_y which give the displacements of nearest-neighboring Cu_I's relative to a Cu_{II}. Right: the vectors $2\delta_x$ and $2\delta_y$ between nearest-neighboring Cu_{II} spins.

$$\mathcal{H}' = -\tfrac{1}{2}\Delta J_1 \sum_{i\in I,\delta_1} S_i^z S_{i+\delta_1}^z - \Delta J_{12} \sum_{i\in II,\delta_{2,1}} S_i^z S_{i+\delta_{2,1}}^z - \tfrac{1}{2}\Delta J_2 \sum_{i\in II,\delta_2} S_i^z S_{i+\delta_2}^z + \tfrac{1}{2}\delta J_1 \sum_{i\in I,\delta_+} (S_i^x S_{i+\delta_+}^y + S_i^y S_{i+\delta_+}^x)$$
$$- \tfrac{1}{2}\delta J_1 \sum_{i\in I,\delta_-} (S_i^x S_{i+\delta_-}^y + S_i^y S_{i+\delta_-}^x) + \delta J_{12} \sum_{i\in II,\delta_x} (S_i^x S_{i+\delta_x}^x - S_i^y S_{i+\delta_x}^y) + \delta J_{12} \sum_{i\in II,\delta_y} (S_i^y S_{i+\delta_y}^y - S_i^x S_{i+\delta_y}^x)$$
$$+ \tfrac{1}{2}\delta J_2 \sum_{i\in II} \sum_{\delta_2 : j=i+\delta_2} [[\mathbf{S}_i\cdot\hat{\delta}_2][\mathbf{S}_j\cdot\hat{\delta}_2] - [\mathbf{S}_i\cdot\hat{e}_2][\mathbf{S}_j\cdot\hat{e}_2]], \tag{9}$$

where we introduce the following sums over the δ's:

$$\delta_x = \pm\tfrac{1}{2}a\hat{x}, \quad \delta_y = \pm\tfrac{1}{2}a\hat{y}, \quad \delta_+ = \pm\tfrac{1}{2}a(\hat{x}+\hat{y}), \quad \delta_- = \pm\tfrac{1}{2}a(\hat{x}-\hat{y}), \tag{10}$$

as shown in Fig. 2. In Eq. (8), $\tilde{J} = J + \tfrac{1}{3}\Delta J$ and similarly for the other J's. Since the anisotropy in the J's is so small (at most of order 10^{-3}), we henceforth drop the tildes.

It is convenient to express the spin components in a coordinate system in which one axis (the ξ axis) lies along the line of the staggered magnetization. Thus we introduce the axes ξ and η which are obtained from x and y by a rotation about the z axis of $\pi/4$. Then

$$S^x = (S^\xi - S^\eta)/\sqrt{2}, \quad S^y = (S^\xi + S^\eta)/\sqrt{2}, \tag{11}$$

so that

$$\mathcal{H}' = -\tfrac{1}{2}\Delta J_1 \sum_{i\in I,\delta_1} S_i^z S_{i+\delta_1}^z - \Delta J_{12} \sum_{i\in I,\delta_{2,1}} S_i^z S_{i+\delta_{2,1}}^z - \tfrac{1}{2}\Delta J_2 \sum_{i\in II,\delta_2} S_i^z S_{i+\delta_2}^z + \tfrac{1}{2}\delta J_1 \sum_{i\in I,\delta_+} (S_i^\xi S_{i+\delta_+}^\xi - S_i^\eta S_{i+\delta_+}^\eta)$$
$$+ \tfrac{1}{2}\delta J_1 \sum_{i\in I,\delta_-} (S_i^\eta S_{i+\delta_-}^\eta - S_i^\xi S_{i+\delta_-}^\xi) - \delta J_{12} \sum_{i\in II,\delta_x} (S_i^\xi S_{i+\delta_x}^\eta + S_i^\eta S_{i+\delta_x}^\xi) + \delta J_{12} \sum_{i\in II,\delta_y} (S_i^\xi S_{i+\delta_y}^\eta + S_i^\eta S_{i+\delta_y}^\xi)$$
$$- \delta J_2 \sum_{i\in e} \sum_{\delta_x : j=i+2\delta_x} (S_i^\xi S_j^\eta + S_i^\eta S_j^\xi) + \delta J_2 \sum_{i\in e} \sum_{\delta_y : j=i+2\delta_y} (S_i^\xi S_j^\eta + S_i^\eta S_j^\xi), \tag{12}$$

where, in the last line, $i\in e$ indicates that the sum is taken over only half the Cu_{II} spins, i.e., those on the e sublattice (see Fig. 1).

III. BOSON HAMILTONIAN

A. Overview of the calculation

Since the Cu_I-Cu_{II} interaction is frustrated, the Cu_I and Cu_{II} sublattices are decoupled within mean-field theory or within harmonic spin-wave theory at zero wave vector. In other words, to calculate the energy gaps at zero wave vector we will need to include fluctuations, as first indicated by Shender.[8] Here, in view of the myriad of terms in the Hamiltonian, we need to proceed in as systematic a way as possible. In the original work of Shender[8] it was found that the effective coupling between sublattices, which depends on fluctuations beyond mean-field theory or beyond harmonic spin-wave theory involved energies of relative order $1/S$ with respect to energies encountered in mean-field theory. Accordingly, here we will calculate all relevant effects in the spin-wave spectrum due to anharmonic perturbations up to first order in $1/S$. Therefore we analyze perturbative contributions at one-loop order. To be more specific, we will introduce the usual Dyson-Maleev boson representation[17] of spin operators, in terms of which anharmonic perturbations involving three (four) boson operators are of relative order $1/\sqrt{S}$ $(1/S)$. This means that we treat four-operator perturbations within first-order perturbation theory and three-operator perturbations within second-order perturbation theory, as was done by Rastelli and Tassi[18] in a similar situation. In technical language, this would be done by keeping all such contributions to the wave vector and energy-dependent self-energy. Since we work to low order, a more naive approach (which is entirely equivalent to calculating the self-energy) is both convenient and easy to follow. In this naive approach one truncates all four operator terms by contracting out pairs of operators in all possible ways. This reproduces exactly the results of the one-loop diagrams obtained by treating the four operator vertices in first order perturbation theory. In addition, we would note that all non-Hermitian terms at order $1/S$ do not contribute to first order energies. So, at order $1/S$ we simply discard non-Hermitian terms. Since the three-operator terms are of interest in producing small gaps, we will follow a calculational method which is strictly correct only at zero wave vector. The fact that in our treatment the small perturbations have the wrong dependence on wave vector is irrelevant because their effect is only nonnegligible very near zero wave vector. To avoid the algebraic complexities due to the fact that the magnetic structure has six sublattices, we simply construct, by the methods mentioned above, the effective quadratic Hamiltonian which includes all the self-energy corrections at order $1/S$. As a check that our calculations are really as consistent as we claim, we verify that the gaps have the expected dependence on the perturbations. In other words, when the perturbations are known to not produce gaps, our calculations reproduce that result. This type of check indicates that, for instance, our treatment of three-operator terms in second-order perturbation theory is consistent with our treatment of four-operator terms in first-order perturbation theory.

B. Transformation to bosons

We make the following Dyson-Maleev transformation[17] to bosons $(a,b,\ldots,f)$:

$$
\begin{aligned}
&S_a^+=\sqrt{2S}a, \quad S_a^-=\sqrt{2S}a^\dagger\phi(a), \quad S_a^\xi=S-a^\dagger a\\
&S_b^+=\sqrt{2S}b^\dagger, \quad S_b^-=\sqrt{2S}\phi(b)b, \quad S_b^\xi=-S+b^\dagger b\\
&S_c^+=\sqrt{2S}c^\dagger, \quad S_c^-=\sqrt{2S}\phi(c)c, \quad S_c^\xi=-S+c^\dagger c\\
&S_d^+=\sqrt{2S}d, \quad S_d^-=\sqrt{2S}d^\dagger\phi(d), \quad S_d^\xi=S-d^\dagger d\\
&S_e^+=\sqrt{2S}e^\dagger, \quad S_e^-=\sqrt{2S}\phi(e)e, \quad S_e^\xi=-S+e^\dagger e\\
&S_f^+=\sqrt{2S}f, \quad S_f^-=\sqrt{2S}f^\dagger\phi(f), \quad S_f^\xi=S-f^\dagger f, \qquad (13)
\end{aligned}
$$

where $S^{\pm}=S^{\eta}\pm iS^{z}$, $\phi(x)=1-x^\dagger x/(2S)$, and we have left the site labels implicit. In bosonic variables the isotropic interaction between spins assumes the form

$$
\begin{aligned}
\mathbf{S}_{ai}\cdot\mathbf{S}_{bj}&=S(a_i^\dagger a_i+b_j^\dagger b_j+a_ib_j+a_i^\dagger b_j^\dagger)\\
&\quad-\tfrac{1}{2}(b_j^\dagger b_jb_ja_i+b_j^\dagger a_i^\dagger a_i^\dagger a_i+2a_i^\dagger a_ib_j^\dagger b_j),\\
\mathbf{S}_{ai}\cdot\mathbf{S}_{ej}&=S(a_i^\dagger a_i+e_j^\dagger e_j+a_ie_j+a_i^\dagger e_j^\dagger)\\
&\quad-\tfrac{1}{2}(e_j^\dagger e_je_ja_i+e_j^\dagger a_i^\dagger a_i^\dagger a_i+2a_i^\dagger a_ie_j^\dagger e_j),\\
\mathbf{S}_{ai}\cdot\mathbf{S}_{fj}&=S(-a_i^\dagger a_i-f_j^\dagger f_j+a_i^\dagger f_j+a_if_j^\dagger)\\
&\quad-\tfrac{1}{2}(a_if_j^\dagger f_j^\dagger f_j+f_ja_i^\dagger a_i^\dagger a_i-2a_i^\dagger a_if_j^\dagger f_j)\\
\mathbf{S}_{bi}\cdot\mathbf{S}_{ej}&=S(-b_i^\dagger b_i-e_j^\dagger e_j+b_i^\dagger e_j+b_ie_j^\dagger)\\
&\quad-\tfrac{1}{2}(b_i^\dagger e_j^\dagger e_je_j+e_j^\dagger b_i^\dagger b_ib_i-2b_i^\dagger b_ie_j^\dagger e_j),\\
\mathbf{S}_{bi}\cdot\mathbf{S}_{fj}&=S(b_i^\dagger b_i+f_j^\dagger f_j+b_i^\dagger f_j^\dagger+b_if_j)\\
&\quad-\tfrac{1}{2}(b_i^\dagger f_j^\dagger f_j^\dagger f_j+f_jb_i^\dagger b_ib_i+2b_i^\dagger b_if_j^\dagger f_j),\\
\mathbf{S}_{ei}\cdot\mathbf{S}_{fj}&=S(e_i^\dagger e_i+f_j^\dagger f_j+e_i^\dagger f_j^\dagger+e_if_j)\\
&\quad-\tfrac{1}{2}(e_i^\dagger f_j^\dagger f_j^\dagger f_j+f_je_i^\dagger e_ie_i+2e_i^\dagger e_if_j^\dagger f_j). \qquad (14)
\end{aligned}
$$

The other interactions can be obtained by appropriate relabeling of boson variables.

The effective bilinear spin-wave Hamiltonian is of the form (see below)

$$
\mathcal{H}=\sum_{\mathbf{q}}\,[A(\mathbf{q})_{\mu\nu}\xi_\mu(\mathbf{q})^\dagger\xi_\nu(\mathbf{q})+\tfrac{1}{2}B(\mathbf{q})_{\mu\nu}\xi_\mu^\dagger(\mathbf{q})\xi_\nu^\dagger(-\mathbf{q})+\tfrac{1}{2}B(\mathbf{q})^*_{\mu\nu}\xi_\mu(\mathbf{q})\xi_\nu(-\mathbf{q})], \qquad (15)
$$

where $\xi_1(\mathbf{q})=a(\mathbf{q})$ and so forth (in order b, c, d, e, and f). Here

$$
\xi_\mu^\dagger(i)=\frac{1}{\sqrt{N_{\mathrm{uc}}}}\sum_{\mathbf{q}}\,e^{i\mathbf{q}\cdot\mathbf{r}_i}\xi_\mu^\dagger(\mathbf{q}), \qquad (16)
$$

where N_{uc} is the number of unit cells.

C. Spin-wave spectrum: General considerations

The transformation to normal mode operators $\tau_k(\mathbf{q})$ is

$$
\begin{aligned}
\xi_i^\dagger(\mathbf{q})&=\sum_j\,P_{ij}(\mathbf{q})^*\tau_j^\dagger(\mathbf{q})+\sum_j\,Q_{ij}(\mathbf{q})^*\tau_j(-\mathbf{q}),\\
\xi_i(-\mathbf{q})&=\sum_j\,Q_{ij}(\mathbf{q})^*\tau_j^\dagger(\mathbf{q})+\sum_j\,P_{ij}(\mathbf{q})^*\tau_j(-\mathbf{q}). \qquad (17)
\end{aligned}
$$

To preserve the commutation relations we require that

$$
\begin{aligned}
\mathbf{P}(\mathbf{q})\mathbf{P}^\dagger(\mathbf{q})-\mathbf{Q}(\mathbf{q})\mathbf{Q}^\dagger(\mathbf{q})&=\mathcal{I},\\
\mathbf{P}(\mathbf{q})\mathbf{Q}^\dagger(\mathbf{q})-\mathbf{Q}(\mathbf{q})\mathbf{P}^\dagger(\mathbf{q})&=0, \qquad (18)
\end{aligned}
$$

where $\mathcal{I}$ is the unit matrix.

The transformation inverse to Eq. (17) is therefore

$$\tau_j^\dagger(\mathbf{q}) = \sum_k P_{kj}(\mathbf{q})\xi_k^\dagger(\mathbf{q}) - \sum_k Q_{kj}(\mathbf{q})\xi_k(-\mathbf{q})$$

$$\tau_j(\mathbf{q}) = -\sum_k Q_{kj}(\mathbf{q})^*\xi_k^\dagger(-\mathbf{q}) + \sum_k P_{kj}(\mathbf{q})^*\xi_k(\mathbf{q}). \tag{19}$$

The equation that determines the normal modes is

$$[\tau_j(\mathbf{q}),\mathcal{H}]_- = \omega_j(\mathbf{q})\tau_j(\mathbf{q}) \tag{20}$$

which gives

$$\begin{bmatrix} \mathbf{A}(\mathbf{q}) & \mathbf{B}(\mathbf{q}) \\ -\mathbf{B}(\mathbf{q}) & -\mathbf{A}(\mathbf{q}) \end{bmatrix} \begin{bmatrix} \mathbf{P}_j(\mathbf{q}) \\ \mathbf{Q}_j(\mathbf{q}) \end{bmatrix} = \omega_j(\mathbf{q}) \begin{bmatrix} \mathbf{P}_j(\mathbf{q}) \\ \mathbf{Q}_j(\mathbf{q}) \end{bmatrix}. \tag{21}$$

where $\mathbf{P}_j$ is the column vector with components $P_{1j}, P_{2j}, \ldots, P_{nj}$ and $\mathbf{P} = [\mathbf{P}_1, \mathbf{P}_2, \ldots \mathbf{P}_n]$ and similarly for the $\mathbf{Q}$'s.

From now on the arguments are always $\mathbf{q}$. Then

$$[\mathbf{A}+\mathbf{B}][\mathbf{P}_j+\mathbf{Q}_j] = \omega_j[\mathbf{P}_j-\mathbf{Q}_j],$$

$$[\mathbf{A}-\mathbf{B}][\mathbf{P}_j-\mathbf{Q}_j] = \omega_j[\mathbf{P}_j+\mathbf{Q}_j]. \tag{22}$$

Therefore

$$[\mathbf{A}+\mathbf{B}][\mathbf{A}-\mathbf{B}][\mathbf{P}_j-\mathbf{Q}_j] = \omega_j^2[\mathbf{P}_j-\mathbf{Q}_j]. \tag{23}$$

Hence, the squares of the spin-wave energies are the eigenvalues of the matrix

$$\mathbf{D}(\mathbf{q}) \equiv [\mathbf{A}(\mathbf{q})+\mathbf{B}(\mathbf{q})] \times [\mathbf{A}(\mathbf{q})-\mathbf{B}(\mathbf{q})]. \tag{24}$$

Roughly speaking the matrices $\mathbf{A}+\mathbf{B}$ and $\mathbf{A}-\mathbf{B}$ reproduce the stiffnesses in the two directions transverse to the sublattice magnetization.

As we shall see later, for the Hamiltonian of the form of $\mathcal{H}_1$ these dynamical matrices assume the form

$$\mathbf{A}(\mathbf{q}) = \begin{bmatrix} a_{11} & a_{12}c_+ & a_{12}c_- & 0 & a_{15}e_x & a_{16}e_x^* \\ a_{12}c_+ & a_{11} & 0 & a_{12}c_- & a_{16}e_y^* & a_{15}e_y \\ a_{12}c_- & 0 & a_{11} & a_{12}c_+ & a_{16}e_y & a_{15}e_y^* \\ 0 & a_{12}c_- & a_{12}c_+ & a_{11} & a_{15}e_x^* & a_{16}e_x \\ a_{15}e_x^* & a_{16}e_y & a_{16}e_y^* & a_{15}e_x & a_{55} & a_{56}\dfrac{c_x+c_y}{2} \\ a_{16}e_x & a_{15}e_y^* & a_{15}e_y & a_{16}e_x^* & a_{56}\dfrac{c_x+c_y}{2} & a_{55} \end{bmatrix} \tag{25a}$$

and

$$\mathbf{B}(\mathbf{q}) = \begin{bmatrix} b_{11} & b_{12}c_+ + 2J_3Sc_z & b_{12}c_- & 0 & b_{15}e_x & b_{16}e_x^* \\ b_{12}c_+ + 2J_3Sc_z & b_{11} & 0 & b_{12}c_- & b_{16}e_y^* & b_{15}e_y \\ b_{12}c_- & 0 & b_{11} & b_{12}c_+ + 2J_3Sc_z & b_{16}e_y & b_{15}e_y^* \\ 0 & b_{12}c_- & b_{12}c_+ + 2J_3Sc_z & b_{11} & b_{15}e_x^* & b_{16}e_x \\ b_{15}e_x^* & b_{16}e_y & b_{16}e_y^* & b_{15}e_x & b_{55} & b_{56}\dfrac{c_x+c_y}{2} \\ b_{16}e_x & b_{15}e_y^* & b_{15}e_y & b_{16}e_x^* & b_{56}\dfrac{c_x+c_y}{2} & b_{55} \end{bmatrix}, \tag{25b}$$

where

$$e_x = \exp(iq_xa/2), \quad e_y = \exp(iq_ya/2), \quad c_x = \cos(q_xa),$$

$$c_y = \cos(q_ya)$$

$$c_+ = \cos[a(q_x+q_y)/2], \quad c_- = \cos[a(q_x-q_y)/2],$$

$$c_z = \cos(q_zc/2). \tag{26}$$

From now on we will analyze the energies of the modes for wave vectors of the form $\mathbf{G}+q_z\hat{z}$, where $\mathbf{G}$ is a reciprocal lattice vector. In that case the matrices $\mathbf{A}$ and $\mathbf{B}$ can be brought into block diagonal form consisting of three 2×2 blocks. The unitary transformation such that $\mathbf{U}^\dagger\mathbf{A}\mathbf{U}$ and

$\mathbf{U}^\dagger\mathbf{B}\mathbf{U}$ are block diagonal depends on $\mathbf{G}$, although, of course, the mode energies do not. For $\mathbf{G}=0$ we have

$$\mathbf{U}=\begin{bmatrix} 1/\sqrt{2} & 0 & 1/2 & 0 & 1/2 & 0 \\ 0 & 1/\sqrt{2} & 1/2 & 0 & -1/2 & 0 \\ 0 & -1/\sqrt{2} & 1/2 & 0 & -1/2 & 0 \\ -1/\sqrt{2} & 0 & 1/2 & 0 & 1/2 & 0 \\ 0 & 0 & 0 & 1/\sqrt{2} & 0 & 1/\sqrt{2} \\ 0 & 0 & 0 & 1/\sqrt{2} & 0 & -1/\sqrt{2} \end{bmatrix}. \tag{27}$$

$\mathbf{U}(\mathbf{G})$ for general $\mathbf{G}$ is given in Appendix A. For $\mathbf{G}=0$ the transformed block-diagonal matrices corresponding to columns 1 and 2, (labeled ''12''), those for columns 3 and 4 (labeled $\sigma=+1$), and those for columns 5 and 6 (labeled $\sigma=-1$) are

$$\mathbf{A}_{12}=\begin{bmatrix} a_{11} & 0 \\ 0 & a_{11} \end{bmatrix}, \tag{28a}$$

$$\mathbf{A}_{\sigma}=\begin{bmatrix} a_{11}+2\sigma a_{12} & \sqrt{2}(a_{15}+\sigma a_{16}) \\ \sqrt{2}(a_{15}+\sigma a_{16}) & a_{55}+\sigma a_{56} \end{bmatrix}, \tag{28b}$$

$$\mathbf{B}_{12}=\begin{bmatrix} b_{11} & 2J_3Sc_z \\ 2J_3Sc_z & b_{11} \end{bmatrix}, \tag{29a}$$

$$\mathbf{B}_{\sigma}=\begin{bmatrix} b_{11}+2\sigma b_{12}+2\sigma J_3Sc_z & \sqrt{2}(b_{15}+\sigma b_{16}) \\ \sqrt{2}(b_{15}+\sigma b_{16}) & b_{55}+\sigma b_{56} \end{bmatrix}. \tag{29b}$$

These results remain valid when $\mathcal{H}_2$ is included, providing it is evaluated at zero wave vector, which, as we have said, is an excellent approximation.

D. Isotropic interactions

For a qualitative understanding of the mode structure we start by considering the results of linearized spin-wave theory when all exchange interactions are isotropic. Then one has

$$a_{11}=4JS+2J_3S, \quad a_{16}=b_{15}=J_{12}S,$$
$$a_{55}=b_{56}=4J_2S, \quad b_{12}=2JS \tag{30}$$

and all the other matrix elements are zero.

In the ''12'' sector, we find two optical modes which are degenerate for all q_z, with

$$(\omega/S)^2=(4J+2J_3)^2-(2J_3c_z)^2\approx 16J^2. \tag{31}$$

Spin-wave interactions and anisotropic exchange interactions will have only negligible effects on these optical modes and accordingly we will generally not discuss these modes any further.[20] In the $\sigma=+1$ sector we find modes with energies

$$(\omega_+^{>}/S)^2=2J_3(1-c_z)[8J+2J_3(1+c_z)]\approx 16JJ_3(1-c_z),$$
$$\omega_+^{<}=0. \tag{32}$$

Finally, the $\sigma=-1$ sector has modes whose energies are

$$(\omega_-^{>}/S)^2=2J_3(1-c_z)[8J+2J_3(1+c_z)]$$
$$\approx 16JJ_3(1-c_z), \quad \omega_-^{<}=0. \tag{33}$$

Note that all modes are gapless at zero wave vector and that for both $\sigma=+1$ and $\sigma=-1$ we have a dispersionless zero frequency mode due to the frustration of the Cu_I-Cu_{II} interaction.

Several aspects of the above results are noteworthy. First of all, as we will see from our calculation of the dynamic structure factor in Sec. VII, the $\sigma=+1$ ($\sigma=-1$) sector corresponds to modes in which the spins move out of (within) the basal plane and therefore we will refer to these modes as out-of-plane (in-plane) modes. (This identification can also be deduced from the way the mode energies depend on the out-of-plane and in-plane anisotropies.) For both out-of-plane and in-plane modes note the existence of a completely gapless mode: when the Cu_I's rotate in phase, they produce zero coupling on the Cu_{II}'s, each plane of which can be rotated with zero cost in energy. The higher-energy out-of-plane and in-plane modes are degenerate because we have not yet included any anisotropy and these modes give rise to the usual twofold degenerate mode of the Cu_I subsystem. Even when more general anisotropic interactions are included, the higher-energy modes remain mostly on the Cu_I's and the lower-energy modes remain mostly on the Cu_{II}'s.

E. Mode energies for general interactions

Here we give the mode energies in terms of the matrix elements of Eq. (25) for general interactions for wave vectors of the form $\mathbf{q}=(0,0,q_z)$. (The eigenvalues, but not the matrices, are invariant under addition of a reciprocal lattice vector $\mathbf{G}$ to $\mathbf{q}$.) To evaluate Eq. (24) within the low-frequency sectors $\sigma=\pm 1$, we record the form of the two by two blocks. Since we need both $\mathbf{A}+\mathbf{B}$ and $\mathbf{A}-\mathbf{B}$, we write

$$[\mathbf{A}+\eta\mathbf{B}]_\sigma=\begin{bmatrix} a_{11}+2\sigma a_{12}+\eta b_{11}+2\sigma\eta J_3Sc_z+2\sigma\eta b_{12} & \sqrt{2}[a_{15}+\sigma a_{16}+\eta b_{15}+\sigma\eta b_{16}] \\ \sqrt{2}[a_{15}+\sigma a_{16}+\eta b_{15}+\sigma\eta b_{16}] & a_{55}+\sigma a_{56}+\eta b_{55}+\sigma\eta b_{56} \end{bmatrix} \tag{34}$$

In evaluating Eq. (24) it is useful to note that in the $\sigma=+1$ sector the matrix element $[\mathbf{A}_{11}+\mathbf{B}_{11}]_+\sim 8JS$ is by far the largest matrix element. Similarly in the $\sigma=-1$ sector $[\mathbf{A}_{11}-\mathbf{B}_{11}]_-\sim 8JS$ is by far the largest matrix element. In either case, then, Eq. (24) gives the squares of the mode energies as the eigenvalues of a matrix (or its transpose) of the form

$$\begin{bmatrix} U & V \\ V & W \end{bmatrix}\begin{bmatrix} u & v \\ u & w \end{bmatrix}, \tag{35}$$

where $\sqrt{Uu}$ dominates all other matrix elements. In that case the eigenvalues are

$$(\omega^>)^2=Uu+2Vv+Wv^2/u,$$
$$(\omega^<)^2=(UW-V^2)(uw-v^2)/(\omega^>)^2. \tag{36}$$

Explicitly, within the sectors $\sigma=\pm 1$, we have

$$U_\sigma=a_{11}+2b_{12}+\sigma(2a_{12}+b_{11})+2J_3Sc_z, \tag{37}$$
$$V_\sigma=\sqrt{2}[a_{15}+b_{16}+\sigma(a_{16}+b_{15})],$$
$$W_\sigma=a_{55}+b_{56}+\sigma(a_{56}+b_{55}),$$
$$u_\sigma=a_{11}-2b_{12}-2J_3Sc_z+\sigma(2a_{12}-b_{11}),$$
$$v_\sigma=\sqrt{2}[a_{15}-b_{16}+\sigma(a_{16}-b_{15})],$$
$$w_\sigma=a_{55}-b_{56}+\sigma(a_{56}-b_{55}).$$

Substituting these evaluations into Eq. (36) [or, if need be, exactly implementing Eq. (24)] gives the four low-energy modes for wave vectors along the $\underline{c}$ direction. Obviously, since the mode energies are derived from a two by two dynamical matrix, we can easily obtain exact expressions for their energies.

IV. NONLINEAR SPIN WAVES

A. 1/*S* corrections to *J*, J_3, and J_2

When we include the effect of spin-wave interactions at order $1/S$ on the Cu_I-Cu_I interactions or on the Cu_{II}-Cu_{II} interactions, we expect to get a simple renormalization. For the exchange interactions between neighbors in the same CuO plane, this effect is well known. As explained above, we decouple the fourth order terms in $\mathbf{S}_{ai}\cdot\mathbf{S}_{bj}$ as

$$-\tfrac{1}{2}[b_j^\dagger b_j b_j a_i+b_j^\dagger a_i^\dagger a_i^\dagger a_i+2a_i^\dagger a_i b_j^\dagger b_j]$$
$$\rightarrow -\langle a_i^\dagger a_i+a_i b_j\rangle[a_i^\dagger a_i+b_j^\dagger b_j+a_i b_j+a_i^\dagger b_j^\dagger] \tag{38}$$

and those in $\mathbf{S}_{ei}\cdot\mathbf{S}_{fj}$ as

$$-\tfrac{1}{2}[e_i^\dagger f_j^\dagger f_j^\dagger f_j+f_j e_i^\dagger e_i e_i+2e_i^\dagger e_i f_j^\dagger f_j]$$
$$\rightarrow -\langle e_i^\dagger e_i+e_i f_j\rangle[e_i^\dagger e_i+f_j^\dagger f_j+e_i f_j+e_i^\dagger f_j^\dagger]. \tag{39}$$

From this result we conclude that J and J_2 should be replaced by Z_cJ and Z_2J_2, respectively, with $Z_c=1-(1/S)\langle a_i^\dagger a_i+a_i b_j\rangle$, where i and j are nearest-neighboring sites on the $\underline{a}$ and $\underline{b}$ sublattices, respectively, and $Z_2=1-(1/S)\langle e_i^\dagger e_i+e_i f_j\rangle$ in a similar notation, so that $Z_2\approx Z_c$ at zero temperature. Z_c has been calculated more accurately than this. (In Ref. 21 the value $Z_c\approx 1.17$ is given.) For J_3 we note that a_i and b_j refer to sites in different CuO planes, in which case $\langle a_i b_j\rangle\approx 0$. So we should replace J_3 by $\tilde{Z}_3J_3$, where

$$\tilde{Z}_3=1-(1/S)\langle a_i^\dagger a_i\rangle, \tag{40}$$

so that $\tilde{Z}_3/2$ is essentially the magnitude of the zero-point staggered spin in the presence of quantum fluctuations. (Thus $\tilde{Z}_3\approx 0.6$ is very different from Z_c.)

B. The effect of spin-wave interactions on J_{12}

Now we discuss the effect of spin-wave interactions on J_{12}, i.e., we consider the Shender interaction.[8] Correctly to order $1/S$ we construct the effective quadratic Hamiltonian by contracting two operators in all possible ways. That is, we replace two operators by the thermal expectation value (indicated by $\langle\cdots\rangle$) of their product. Applying this procedure to the relevant terms in Eq. (14) we obtain the effective interactions between a Cu_I spin i on sublattice $\underline{a}$ and nearest neighboring Cu_{II} spins as

$$V_{ae}/J_{12}=a_i^\dagger a_i(S-\langle a_i^\dagger e_j^+\rangle-\langle e_j^\dagger e_j\rangle)+e_j^\dagger e_j(S-\langle e_j a_i\rangle$$
$$-\langle a_i^\dagger a_i\rangle)+a_i e_j(S-\langle e_j^\dagger e_j\rangle-\langle a_i^\dagger e_j^\dagger\rangle)$$
$$+a_i^\dagger e_j^\dagger(S-\langle a_i^\dagger a_i\rangle-\langle a_i e_j\rangle), \tag{41}$$

$$V_{af}/J_{12}=a_i^\dagger a_i(-S-\langle a_i^\dagger f_j\rangle+\langle f_j^\dagger f_j\rangle)+f_j^\dagger f_j(-S-\langle a_i f_j^\dagger\rangle$$
$$+\langle a_i^\dagger a_i\rangle)+a_i^\dagger f_j(S-\langle a_i^\dagger a_i\rangle+\langle a_i f_j^\dagger\rangle)$$
$$+f_j^\dagger a_i(S-\langle f_j^\dagger f_j\rangle+\langle a_i^\dagger f_j\rangle). \tag{42}$$

Here to leading order in $1/S$ it suffices to evaluate the various expectation values with respect to the original quadratic Hamiltonian. At quadratic order we have symmetry such that $\langle a_i^\dagger e_j^\dagger\rangle=\langle a_i e_j\rangle,\langle e_j^\dagger e_j\rangle=\langle f_j^\dagger f_j\rangle$, etc. We define

$$J_{12}^{(1)}S/J_{12}=S+\langle a_i f_j^\dagger\rangle-\langle a_i^\dagger a_i\rangle,$$
$$J_{12}^{(2)}S/J_{12}=S+\langle a_i f_j^\dagger\rangle-\langle f_j^\dagger f_j\rangle,$$
$$J_{12}^{(3)}S/J_{12}=S-\langle a_i e_j\rangle-\langle a_i^\dagger a_i\rangle,$$
$$J_{12}^{(4)}S/J_{12}=S-\langle a_i e_j\rangle-\langle e_j^\dagger e_j\rangle. \tag{43}$$

Note that $J_{12}^{(3)}-J_{12}^{(4)}=J_{12}^{(1)}-J_{12}^{(2)}$. Then

$$V_{ae}=J_{12}^{(4)}Sa_i^\dagger a_i+J_{12}^{(3)}Se_j^\dagger e_j+J_{12}^{(4)}Sa_i e_j+J_{12}^{(3)}Sa_i^\dagger e_j^\dagger,$$
$$V_{af}=-J_{12}^{(2)}Sa_i^\dagger a_i-J_{12}^{(1)}Sf_j^\dagger f_j+J_{12}^{(1)}Sa_i^\dagger f_j+J_{12}^{(2)}Sf_j^\dagger a_i. \tag{44}$$

Since we only work to order $1/S$, we keep only the Hermitian part of these perturbations:

$$V_{ae}=J_{12}^{(4)}Sa_i^\dagger a_i+J_{12}^{(3)}Se_j^\dagger e_j+J_{12}^{(34)}S(a_ie_j+a_i^\dagger e_j^\dagger),$$
$$V_{af}=-J_{12}^{(2)}Sa_i^\dagger a_i-J_{12}^{(1)}Sf_j^\dagger f_j+J_{12}^{(12)}S(a_i^\dagger f_j+f_j^\dagger a_i), \quad (45)$$

where

$$J_{12}^{(12)}=\tfrac{1}{2}[J_{12}^{(1)}+J_{12}^{(2)}], \quad J_{12}^{(34)}=\tfrac{1}{2}[J_{12}^{(3)}+J_{12}^{(4)}]. \quad (46)$$

As it turns out, the energies of the modes we study depend only on the single parameter

$$\alpha=(J_{12}^{(4)}-J_{12}^{(2)})S=(J_{12}^{(3)}-J_{12}^{(1)})S=-J_{12}(\langle a_ie_j\rangle+\langle a_if_j^\dagger\rangle). \quad (47)$$

Note that the parameter δ in Ref. 5 is $\delta=\alpha/S$. We evaluate this parameter in Appendix B and find

$$\alpha=C_\alpha J_{12}^2/J, \quad (48)$$

where C_α is a numerical factor which we found to be 0.1686. The anharmonic effects of Eq. (45) give rise to contributions to the dynamical matrix of

$$\delta a_{11}=\alpha,$$
$$\delta a_{16}=J_{12}^{(12)}S-J_{12}S,$$
$$\delta a_{55}=2\alpha,$$
$$\delta b_{15}=J_{12}^{(34)}S-J_{12}S. \quad (49)$$

It is known[8,22,23] that in simpler problems these anharmonic effects give rise *at zero momentum* to effective biquadratic exchange interactions between sublattices which otherwise are frustrated in harmonic theory. To emphasize this point we treat a biquadratic interaction between nearest Cu_I-Cu_{II} neighbors (in the plane) which is of the form

$$H_{BQ}=-\frac{j_{BQ}}{S^2}\sum_{i\in II}\sum_{\delta_{2,1}}(\mathbf{S}_i\cdot\mathbf{S}_{i+\delta_{2,1}})^2. \quad (50)$$

Then the contributions to the dynamical matrix are

$$\delta a_{11}=4j_{BQ}S, \quad (51)$$
$$\delta a_{16}=-2j_{BQ}S,$$
$$\delta a_{55}=8j_{BQ}S,$$
$$\delta b_{15}=2j_{BQ}S.$$

Then using Eqs. (37) and (36) we find the mode energies at zero transverse wave vector (for large J) are now

$$(\omega_\sigma^>)^2=8JS[\alpha+4j_{BQ}S+2J_3S(1-c_z)]$$
$$\equiv 8JS[\alpha_{\rm eff}+2J_3S(1-c_z)],$$
$$(\omega_\sigma^<)^2=\frac{4\alpha_{\rm eff}J_3S(1-c_z)(8J_2S+\alpha_{\rm eff})}{\alpha_{\rm eff}+2J_3S(1-c_z)}, \quad (52)$$

where

$$\alpha_{\rm eff}=\alpha+4j_{BQ}S. \quad (53)$$

These results demonstrate that the Shender interaction does mimic a biquadratic exchange interaction at long wave length. However, in view of the relation for spin 1/2 that $(\mathbf{S}_i\cdot\mathbf{S}_j)^2=\frac{3}{16}-\frac{1}{2}\mathbf{S}_i\cdot\mathbf{S}_j$, a biquadratic exchange interaction between two spins 1/2 is equivalent to a Heisenberg exchange interaction, and we may therefore assume that j_{BQ} vanishes.

As before, there is degeneracy between in-plane and out-of-plane energies because we have not yet included anisotropy. However, by taking into account spin-wave interactions we now have the mode structure one would expect for an isotropic antiferromagnet: We have a doubly degenerate zero energy Goldstone mode at zero wave vector, and doubly degenerate nonzero energy modes for nonzero wave vector as shown in the right-hand panel of Fig. 3. The quantum gap in the optical mode $\omega_\sigma^>$ at zero wave vector has been obtained for a number of other frustrated systems in several theoretical studies[24,22,23] beginning with the work of Shender.[8] However, because we have two subsystems which order at different temperatures, the emergence of this gap has a very unique signature not present in other experimental systems studied up to now.[9]

V. INCLUSION OF ANISOTROPIES

A. Out-of-plane exchange anisotropy

To obtain the correct energy gaps at zero wave vector we must add the anisotropy due to anisotropic exchange interactions. (Since we are dealing with spin 1/2's, there can be no single ion anisotropy.) In this subsection we include out-of-plane exchange anisotropy. This part of the anisotropic exchange energy between sublattices $\underline{a}$ and $\underline{b}$ of the Cu_I's is given as

$$V_{ab}\equiv-\Delta J_1\sum_{i\in a,j\in b}S_{ai}^zS_{bj}^z\Delta_{ij}, \quad (54)$$

where Δ_{ij} is defined so as to implement the nearest neighbor restriction. Thus, neglecting anharmonicity, we write

$$V_{ab}=\tfrac{1}{4}\Delta J_1\sum_{i\in a,j\in b}[S_{ai}^+-S_{ai}^-][S_{bj}^+-S_{bj}^-]\Delta_{ij}$$
$$=\tfrac{1}{2}\Delta J_1S\sum_{i\in a,j\in b}(a_i-a_i^\dagger)(b_j^\dagger-b_j)\Delta_{ij}$$
$$=\Delta J_1S\sum_{\mathbf{q}}[a^\dagger(\mathbf{q})b(\mathbf{q})+b^\dagger(\mathbf{q})a(\mathbf{q})-a^\dagger(\mathbf{q})b^\dagger(-\mathbf{q})$$
$$-a(\mathbf{q})b(-\mathbf{q})]c_+. \quad (55)$$

This result allows us to identify the contribution to the parameters of the dynamical matrix introduced in Eq. (25) as

$$\delta a_{12}=\Delta J_1S, \quad \delta b_{12}=-\Delta J_1S, \quad (56)$$

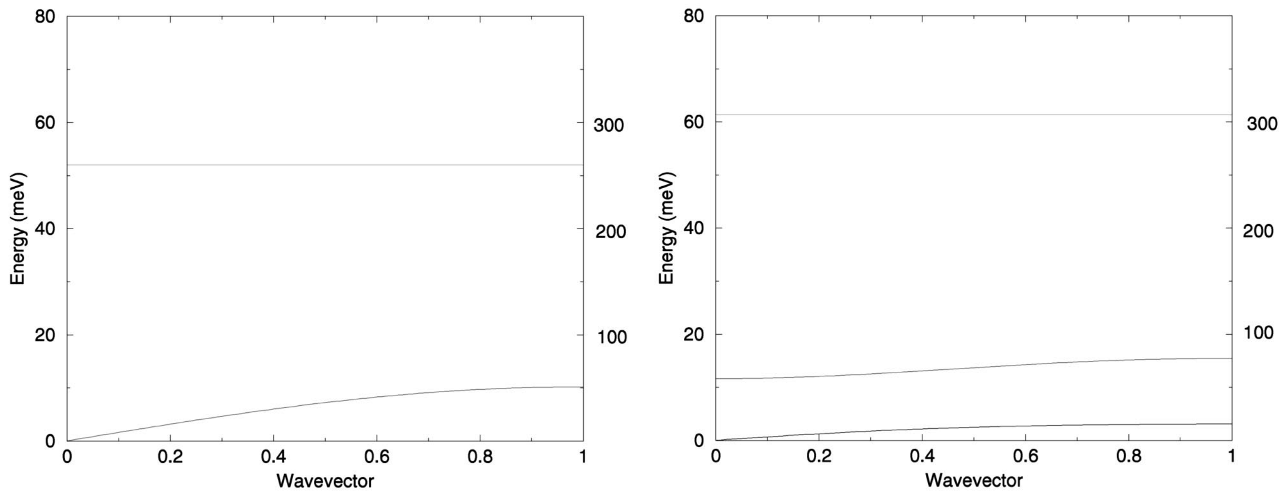

FIG. 3. Spin-wave spectrum for wave vector $=q_z c/(2\pi)$ along the $\underline{c}$ direction in the absence of anisotropy. Each mode is twofold degenerate. The left-hand scale applies to the lower modes and the right-hand scale applies to the optical mode. Left: without spin-wave interactions. In this case one mode has zero energy for arbitrary wave vector in the $\underline{c}$ direction. Right: with spin-wave interactions. In the presence of easy plane anisotropy, the twofold degeneracy is removed and only one mode (corresponding to rotation within the easy plane) is gapless at zero wave vector. When the fourfold in-plane anisotropy is also included there are no gapless modes.

without having to explicitly consider the other Cu_I-Cu_I interactions.

Next we consider the out-of-plane anisotropy of the Cu_I-Cu_{II} interactions. From the form of Eq. (25) we see that we only need construct the a-e and a-f interactions. For the a-e interaction we have

$$\begin{aligned} V_{ae} &= -\Delta J_{12} \sum_{i\in a, j\in e} S^z_{ai} S^z_{ej} \Delta_{ij} \\ &= \tfrac{1}{4}\Delta J_{12} \sum_{i\in a, j\in e} (S^+_{ai} - S^-_{ai})(S^+_{ej} - S^-_{ej})\Delta_{ij} \\ &= \tfrac{1}{2}\Delta J_{12} S \sum_{a\in i, j\in e} (a_i - a_i^\dagger)(e_j^\dagger - e_j)\Delta_{ij} \\ &= \tfrac{1}{2}\Delta J_{12} S \sum_{\mathbf{q}} [a(\mathbf{q}) e^\dagger(\mathbf{q}) e^{i\mathbf{q}\cdot(\mathbf{r}_e - \mathbf{r}_a)} \\ &\quad - a^\dagger(-\mathbf{q}) e^\dagger(\mathbf{q}) e^{i\mathbf{q}\cdot(\mathbf{r}_e - \mathbf{r}_a)} + \text{H.c.}] \\ &= \tfrac{1}{2}\Delta J_{12} S \sum_{\mathbf{q}} [a(\mathbf{q}) e^\dagger(\mathbf{q}) e_x^* - a^\dagger(-\mathbf{q}) e^\dagger(\mathbf{q}) e_x^* \\ &\quad + a^\dagger(\mathbf{q}) e(\mathbf{q}) e_x - a(-\mathbf{q}) e(\mathbf{q}) e_x], \end{aligned} \quad (57)$$

which gives a contribution to the dynamical matrix with

$$\delta a_{15} = \tfrac{1}{2}\Delta J_{12} S, \quad \delta b_{15} = -\tfrac{1}{2}\Delta J_{12} S. \quad (58)$$

Similarly

$$\begin{aligned} V_{af} &= \tfrac{1}{2}\Delta J_{12} S \sum_{\mathbf{q}} [a(\mathbf{q}) f(-\mathbf{q}) e_x - a^\dagger(\mathbf{q}) f(\mathbf{q}) e_x^* \\ &\quad + a^\dagger(\mathbf{q}) f^\dagger(-\mathbf{q}) e_x^* - a(\mathbf{q}) f^\dagger(\mathbf{q}) e_x], \end{aligned} \quad (59)$$

from which we deduce that

$$\delta a_{16} = -\tfrac{1}{2}\Delta J_{12} S, \quad \delta b_{16} = \tfrac{1}{2}\Delta J_{12} S. \quad (60)$$

Finally we include the out-of-plane anisotropy of the Cu_{II}-Cu_{II} interactions. Thus

$$\begin{aligned} V_{ef} &= -\Delta J_2 \sum_{i\in e, j\in f} S^z_{ei} S^z_{fj} \Delta_{ij} \\ &= \tfrac{1}{4}\Delta J_2 \sum_{i\in e, j\in f} [S^+_{ei} - S^-_{ei}][S^+_{fj} - S^-_{fj}]\Delta_{ij} \\ &= \tfrac{1}{2}\Delta J_2 S \sum_{i\in e, j\in f} (e_i^\dagger - e_i)(f_j - f_j^\dagger)\Delta_{ij} \\ &= \Delta J_2 S \sum_{\mathbf{q}} [e^\dagger(\mathbf{q}) f(\mathbf{q}) - e^\dagger(\mathbf{q}) f^\dagger(-\mathbf{q}) - e(\mathbf{q}) f(-\mathbf{q}) \\ &\quad + f^\dagger(\mathbf{q}) e(\mathbf{q})][c_x + c_y], \end{aligned} \quad (61)$$

which leads to

$$\delta a_{56} = 2\Delta J_2 S, \quad \delta b_{56} = -2\Delta J_2 S. \quad (62)$$

The renormalization (at order $1/S$) of the out-of-plane anisotropy is accomplished by replacing $\sqrt{J\Delta J_1}$ by $Z_g\sqrt{J\Delta J_1}$.[25]

It is instructive to see the influence of this anisotropy on the gaps at zero wave vector. Referring to Eq. (36) we see that the high energy mode gap due to the Shender fluctuation term, causes Uu to be nonzero. To check for gaps in the mode energies $\omega_\sigma^<$ at zero wave vector it suffices to consider the quantity

$$\begin{aligned} \Lambda &\equiv uw - v^2 \\ &= [2\Delta J_1 S(1+\sigma) + \alpha][2\alpha + 2\Delta J_2 S(1+\sigma)] \\ &\quad - [-\sqrt{2}\sigma\alpha]^2. \end{aligned} \quad (63)$$

When we turn off both out-of-plane anisotropies ΔJ_1 and ΔJ_2, the two modes $\omega_\sigma^<$ are gapless. When we allow the out-of-plane anisotropy to be nonzero, we clearly introduce a gap (Λ is nonzero) in the out-of-plane ($\sigma=1$) sector but not in the in-plane ($\sigma=-1$) sector. This result follows from the fact that the spins can still undergo a global rotation within the easy plane at no cost in energy. Hence we still have a single Goldstone mode with zero energy at zero wave vector. In order for this mode to have a gap, we have to take account of effects which lead to a fourfold in-plane anisotropy which we consider in the next subsection.

B. In-plane exchange anisotropy

1. Cu_I-Cu_I interactions

In this subsection we discuss the effects of the in-plane anisotropy of the Cu_I-Cu_I exchange interactions. First of all, note that this perturbation is extremely weak. It gives rise to an effective fourfold anisotropy. This very small fourfold anisotropy only has a non-negligible effect within the low-frequency sector and even there only at zero wave vector. The Hamiltonian describing the in-plane anisotropy of the Cu_I-Cu_I interactions is

$$V_{\rm in}=\delta J_1\sum_{i\in a,d;\delta}\sigma(\delta)(S_i^\xi S_{i+\delta}^\xi-S_i^\eta S_{i+\delta}^\eta), \qquad (64)$$

where $j=i+\delta$, δ is summed over four values (the two δ_+'s and the two δ_-'s), and $\sigma(\delta_\pm)=\pm 1$. Then

$$\begin{aligned}V_{\rm in}&=\delta J_1\sum_{i\in a,d;\delta}\sigma(\delta)\{(S-\alpha_i^\dagger\alpha_i)(-S+\beta_j^\dagger\beta_j)\\&\quad-\tfrac14(2S)[\alpha_i+\alpha_i^\dagger\phi(\alpha_i)][\beta_j^\dagger+\phi(\beta_i)\beta_j]\}\\&=\delta J_1\sum_{i\in a,d;\delta}\sigma(\delta)\Big(-\alpha_i^\dagger\alpha_i\beta_j^\dagger\beta_j-\tfrac12 S[\alpha_i+\alpha_i^\dagger][\beta_j^\dagger+\beta_j]\\&\quad+\tfrac14\alpha_i^\dagger\alpha_i^\dagger\alpha_i(\beta_j^\dagger+\beta_j)+\tfrac14(\alpha_i^\dagger+\alpha_i)\beta_j^\dagger\beta_j\beta_j\\&\quad-\frac{1}{8S}\alpha_i^\dagger\alpha_i^\dagger\alpha_i\beta_j^\dagger\beta_j\beta_j\Big),\end{aligned} \qquad (65)$$

where $\alpha_i=a$ if site i is an a site and $\alpha_i=d$ if i is a d site, and similarly for β_j. We write

$$V_{\rm in}=V_{2,\rm in}+V_{4,\rm in}+V_{6,\rm in}, \qquad (66)$$

where the subscript 2 (4 or 6) indicates terms quadratic (fourth or sixth) order in boson operators. Since we work systematically to first order in $1/S$, we neglect $V_{6,\rm in}$. Also

$$\begin{aligned}V_{2,\rm in}&=-\tfrac12\delta J_1 S\sum_{i\delta}\sigma(\delta)(\alpha_i+\alpha_i^\dagger)(\beta_j+\beta_j^\dagger)\\&=-\delta J_1 S\sum_{\delta,\mathbf{k}}\{[a^\dagger(\mathbf{k})+a(-\mathbf{k})][b(\mathbf{k})+b^\dagger(-\mathbf{k})]\\&\quad+[d^\dagger(\mathbf{k})+d(-\mathbf{k})][c(\mathbf{k})+c^\dagger(-\mathbf{k})]\}c_+\\&\quad+\delta J_1 S\sum_{\delta,\mathbf{k}}\{[a^\dagger(\mathbf{k})+a(-\mathbf{k})][c(\mathbf{k})+c^\dagger(-\mathbf{k})]\\&\quad+[d^\dagger(\mathbf{k})+d(-\mathbf{k})][b(\mathbf{k})+b^\dagger(-\mathbf{k})]\}c_-\end{aligned} \qquad (67)$$

and

$$\begin{aligned}V_{4,\rm in}&=\delta J_1\sum_{i\in a,d;\delta}\sigma(\delta)[-\alpha_i^\dagger\alpha_i\beta_j^\dagger\beta_j+\tfrac14\alpha_i^\dagger\alpha_i^\dagger\alpha_i(\beta_j^\dagger+\beta_j)\\&\quad+\tfrac14(\alpha_i^\dagger+\alpha_i)\beta_j^\dagger\beta_j\beta_j].\end{aligned} \qquad (68)$$

We now consider the effect of $V_{2,\rm in}$ on the spectrum for $k_x=k_y=0$, so that $c_+=c_-=1$. In this case because the perturbation is proportional to $b-c$ or to $b^\dagger-c^\dagger$, one sees that $V_{2,\rm in}$ only couples to the optical mode sector. Accordingly, we do not consider $V_{2,\rm in}$ any further.

We expect that this in-plane anisotropy should give rise to a macroscopic fourfold anisotropy. In order to obtain this anisotropy we must include anharmonic effects at relative order $1/S$. Now we decouple the four operator terms into quadratic terms times averages of the remaining quadratic factors. This calculation is done in Appendix C. In that calculation we naturally drop all contributions to the optical mode sector and of the rest keep only terms which have an effect on the mode energies at zero wave vector. The result is that contributions to the dynamical matrices due to the in-plane Cu_I-Cu_I interactions yield

$$\delta a_{11}=16C_2\tau, \qquad (69a)$$

$$\delta a_{12}=-4(6C_2-C_{2c}-4C_{2b})\tau, \qquad (69b)$$

$$\delta b_{11}=8C_{2c}\tau, \qquad (69c)$$

$$\delta b_{12}=-16C_{2b}\tau, \qquad (69d)$$

where $\tau\equiv(\delta J_1)^2/J$ and the C's are lattice sums defined in Eq. (C19) of Appendix C. It turns out that because τ is so small, the only evaluation we need is that $C_2=0.01$. Note that the contributions in Eq. (69) are of relative order $1/S$ which is consistent with the fact that they represent the effect of quantum fluctuations. The fact that they represent a modification in the zero-point energy is reflected by the appearance of the factor $C_2\ll 1$.

2. Cu_I-Cu_{II} interactions

Next we deal with the in-plane anisotropy of the Cu_I-Cu_{II} interactions. The terms in Eq. (12) involving δJ_{12} are

$$\begin{aligned}V_{\delta J_{12}}&=-\delta J_{12}\sum_{i\in \mathrm{II},\delta_x}[S_i^\xi S_{i+\delta_x}^\eta+S_i^\eta S_{i+\delta_x}^\xi]\\&\quad+\delta J_{12}\sum_{i\in\mathrm{II},\delta_y}[S_i^\xi S_{i+\delta_y}^\eta+S_i^\eta S_{i+\delta_y}^\xi].\end{aligned} \qquad (70)$$

In terms of boson operators this is

$$V_{\delta J_{12}}=\delta J_{12}\sqrt{S/2}\sum_i\{[S-e_i^{\dagger}e_i][a_{i+x}+a_{i+x}^{\dagger}\phi(a_{i+x})+d_{i-x}+d_{i-x}^{\dagger}\phi(d_{i-x})]+[e_i^{\dagger}+\phi(e_i)e_i](-2S+a_{i+x}^{\dagger}a_{i+x}+d_{i-x}^{\dagger}d_{i-x})\}$$
$$-\delta J_{12}\sqrt{S/2}\sum_i\{[S-e_i^{\dagger}e_i][b_{i-y}^{\dagger}+\phi(b_{i-y})b_{i-y}+c_{i+y}^{\dagger}+\phi(c_{i+y})c_{i+y}]+[e_i^{\dagger}+\phi(e_i)e_i](2S-b_{i-y}^{\dagger}b_{i-y}-c_{i+y}^{\dagger}c_{i+y})\}$$
$$+\delta J_{12}\sqrt{S/2}\sum_i\{[-S+f_i^{\dagger}f_i][a_{i-x}+a_{i-x}^{\dagger}\phi(a_{i-x})+d_{i+x}+d_{i+x}^{\dagger}\phi(d_{i+x})]$$
$$+[f_i+f_i^{\dagger}\phi(f_i)](-2S+a_{i-x}^{\dagger}a_{i-x}+d_{i+x}^{\dagger}d_{i+x})\}-\delta J_{12}\sqrt{S/2}\sum_i\{[-S+f_i^{\dagger}f_i][b_{i+y}^{\dagger}+\phi(b_{i+y})b_{i+y}+c_{i-y}^{\dagger}$$
$$+\phi(c_{i-y})c_{i-y}]+[f_i+f_i^{\dagger}\phi(f_i)](2S-b_{i+y}^{\dagger}b_{i+y}-c_{i-y}^{\dagger}c_{i-y})\}. \tag{71}$$

This perturbation contains terms linear and terms cubic in the boson operators. The linear terms and (at relative order $1/S$) the cubic terms will shift the equilibrium so that the boson operators are modified as

$$e_i\to e_i+s,\quad f_i\to f_i+s,\quad a_i\to a_i+t,\quad b_i\to b_i+t,$$
$$c_i\to c_i+t,\quad d_i\to d_i+t. \tag{72}$$

These shifts are evaluated in Appendix D, where we find that (to leading order in $1/S$)

$$s=\frac{4\,\delta J_{12}\sqrt{S/2}}{8J_2},$$

$$t=-\frac{2J_{12}s}{8J+4J_3}=-\frac{J_{12}\delta J_{12}\sqrt{S/2}}{J_2(8J+4J_3)}. \tag{73}$$

These are the expected results. As one sees from Eq. (12), the perpendicular field acting on an e spin is $4\delta J_{12}S$ in the positive η direction, so that the perpendicular moment of the e spin is $\Delta S_e=4\,\delta J_{12}S\chi_{\rm II}=4\,\delta J_{12}S/(8J_2)$, which agrees with $\sqrt{2S}s$ when Eq. (73) is used. Further, due to the isotropic exchange, the field acting on an a spin is $2J_{12}\Delta S_e=J_{12}\delta J_{12}S/J_2$ in the negative η direction. Thus $\Delta S_a=-[J_{12}\delta J_{12}S/J_2]\chi_{\rm I}=-[J_{12}\delta J_{12}S/J_2]/[8J+4J_3]$, which agrees with $\sqrt{2S}t$ when Eq. (73) is used. Note that ΔS_e and ΔS_a are both of order S, a result which indicates that the effects here are completely classical.

To determine the effect of $V_{\delta J_{12}}$ on the spin-wave spectrum we need to construct the effective quadratic Hamiltonian, which results from introducing shifts into anharmonic terms. This is done in Appendix D. When we insert these shifts into the cubic terms of $V_{\delta J_{12}}$ we ignore t in comparison to s because $J\gg J_{12}$. Thereby we get contributions to the dynamical matrix of

$$\delta a_{55}=\delta a_{11}=2\,\delta b_{55}=\delta J_{12}^2S/J_2\equiv\zeta S,$$

$$\delta a_{16}=-\delta a_{15}=\delta b_{16}=-\delta b_{15}=\tfrac{1}{4}\zeta S. \tag{74}$$

We also insert these shifts into the four operator terms of the isotropic Hamiltonian. As before we only keep terms arising from replacing two $Cu_{\rm II}$ operators by $\langle e\rangle$. The magnitude of other terms, e.g., $Cu_{\rm I}$-$Cu_{\rm I}$ quartic terms when $Cu_{\rm I}$ shifts $\langle a\rangle$ are kept, are shown in Appendix D to be much smaller than those we have kept. The result of the calculation in Appendix D is that we get the contributions to the dynamical matrix of

$$\delta a_{55}=-\zeta S,\quad \delta b_{55}=-\tfrac{1}{4}\zeta S,$$

$$\delta a_{56}=-\tfrac{3}{4}\zeta S,\quad \delta b_{56}=-\zeta S. \tag{75}$$

Note that these perturbative contributions from the $Cu_{\rm I}$-$Cu_{\rm II}$ in-plane anisotropy, are proportional to S, unlike the case for the other in-plane anisotropies. This indicates that the effect of δJ_{12} (which we called J_{pd} previously[3,4]), is a classical effect which already appeared within mean field theory.[3,4] The other in-plane anisotropies only have an effect when we consider fluctuations. However, since the effect of δJ_{12} is rather small, we do not consider the effects of fluctuation corrections to it.

3. Cu_{II}-Cu_{II} *intraplanar interactions*

Here we consider the in-plane anisotropy of the interactions between pairs of $Cu_{\rm II}$ spins in the same plane. Their interaction is

$$V=-\delta J_2\sum_{i\in e}\left[\sum_{\delta_x:j=i+2\delta_x}(S_i^\xi S_j^\eta+S_i^\eta S_j^\xi)+\sum_{\delta_y:j=i+2\delta_y}(S_i^\xi S_j^\eta+S_i^\eta S_j^\xi)\right]$$

$$=-\delta J_2\sqrt{\frac{S}{2}}\sum_{i,\delta_x}\{(S-e_i^\dagger e_i)[f_j+f_j^\dagger\phi(f_j)]+[e_i^\dagger+\phi(e_i)e_i](-S+f_j^\dagger f_j)\}$$

$$+\delta J_2\sqrt{\frac{S}{2}}\sum_{i,\delta_y}\{(S-e_i^\dagger e_i)[f_j+f_j^\dagger\phi(f_j)]+[e_i^\dagger+\phi(e_i)e_i](-S+f_j^\dagger f_j)\}$$

$$=\delta J_2\sqrt{S/2}\left[\sum_{i,\delta_x}[e_i^\dagger e_i(f_j+f_j^\dagger)-(e_i^\dagger+e_i)f_j^\dagger f_j]-\sum_{i,\delta_y}[e_i^\dagger e_i(f_j+f_j^\dagger)-(e_i^\dagger+e_i)f_j^\dagger f_j]\right]$$

$$=\delta J_2\sqrt{\frac{8S}{N}}\sum_{\mathbf{q},\mathbf{k}}\rho(\mathbf{k})\{[f(\mathbf{k})+f^\dagger(-\mathbf{k})]e^\dagger(\mathbf{q})e(\mathbf{q}-\mathbf{k})-[e(\mathbf{k})+e^\dagger(-\mathbf{k})]f^\dagger(\mathbf{q})f(\mathbf{q}-\mathbf{k})\},\tag{76}$$

where

$$\rho(\mathbf{k})=\frac{1}{2}[\cos(ak_x)-\cos(ak_y)].\tag{77}$$

This Hamiltonian is treated in Appendix E, where the additional contributions to the spin-wave matrices (at $q_z=0$) are found to be

$$\delta a_{55}=-16[\delta J_2^2/J_2][2C_{2a}+C_{2b}]\equiv-16\xi[2C_{2a}+C_{2b}],$$

$$\delta a_{56}=16\xi[2C_{2a}-C_{2b}]\tag{78}$$

and

$$\delta b_{55}=-16\xi C_{2b},\quad \delta b_{56}=48\xi C_{2b},\tag{79}$$

where C_{2a} and C_{2b} are lattice sums defined in Appendix C.

It is interesting to note that apart from a minus sign, these results are exactly the same as in Yildirim *et al.*[13] This difference in sign is to be expected because the Cu_{II}'s are oriented in a hard direction with respect to only Cu_{II}-Cu_{II} interactions. Consequently, this term tends to decrease the gap.

4. Cu_{II}-Cu_{II} interplanar interactions

Here we consider the effect of interactions between a pair of Cu_{II} spins in adjacent planes. The situation we consider is shown in the left panel of Fig. 4, where one sees that the isotropic component of the Cu_{II}-Cu_{II} interplanar interaction is frustrated. To describe the anisotropy of this interaction we introduce the principal axes (shown in the right panel of Fig. 4) as follows:

$$\hat{n}_1=2^{-1/2}(-\hat{x}+\hat{y}),\quad \hat{n}_2=2^{-1/2}(\hat{x}+\hat{y})\cos\psi+\hat{z}\sin\psi,$$

$$\hat{n}_3=2^{-1/2}(\hat{x}+\hat{y})\sin\psi-\hat{z}\cos\psi.\tag{80}$$

The angle ψ is not fixed by symmetry. We then write the anisotropic Cu_{II}-Cu_{II} interaction $\mathcal{H}_{ij}^{\text{II-II}}$ between nearest-neighboring spins i and j in adjacent planes as[10]

$$\mathcal{H}_{ij}^{\text{II-II}}=\sum_{k=1}^{3}K_k[\mathbf{S}_i\cdot\hat{n}_k^{(ij)}][\mathbf{S}_j\cdot\hat{n}_k^{(ij)}],\tag{81}$$

where $\hat{n}_k^{(ij)}$ is the kth principal axes for the pair ij which can be obtained from the right panel of Fig. 4, by a rotation of coordinates, if necessary, and K_k is the associated principal value of the exchange tensor. The contributions of this interaction to the dynamical matrix are evaluated for $q_x=q_y=0$ in Appendix F as

$$\delta a_{55}=\delta a_{66}=4(K_1-K_2c^2-K_3s^2)S+2(K_1+K_3c^2+K_2s^2)Sc_z,\tag{82a}$$

$$\delta a_{56}=\delta a_{65}=2(K_2-K_3)(c^2-s^2)Sc_z,\tag{82b}$$

$$\delta b_{55}=\delta b_{66}=2(K_1-K_2s^2-K_3c^2)Sc_z,\tag{82c}$$

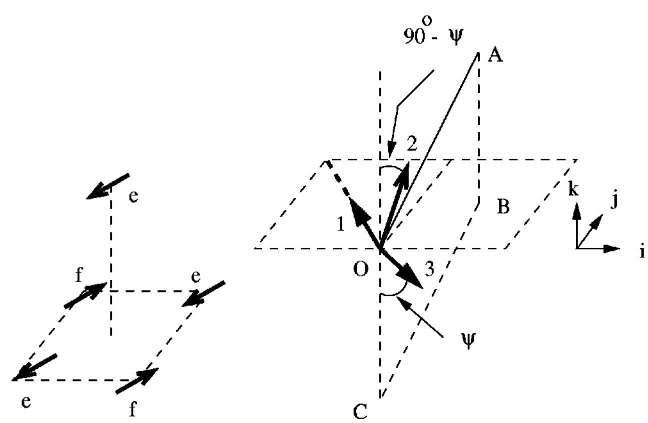

FIG. 4. Interplanar Cu_{II}-Cu_{II} interactions. Left: a plaquette of Cu_{II} spins in one plane with a Cu_{II} neighbor in the adjacent plane over the center of the plaquette such that the isotropic Cu_{II}-Cu_{II} interaction is frustrated. Right: The principal axes for the exchange tensor of a spin in the *e* sublattice at O with a spin in the *e* sublattice at A. The directions of the axes are given in Eq. (80). The axes for the interactions of the spin at A with other spins in the lower plane can be obtained by a rotation of coordinates.

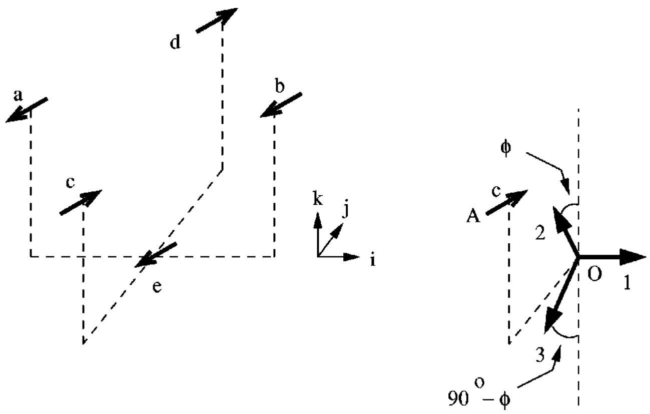

FIG. 5. Interplanar Cu_I-Cu_{II} interactions. Left: a plaquette of Cu_I spins in one plane with a Cu_{II} neighbor in the adjacent plane below the center of the plaquette such that the isotropic Cu_I-Cu_{II} interaction is frustrated. Right: The principal axes for the exchange tensor of a spin in the *e* sublattice at O with a spin in the *c* sublattice at A. The directions of the axes are given in Eq. (80). The axes for the interactions of other pairs of Cu_I-Cu_{II} nearest neighbors in adjacent planes can be obtained by a rotation of coordinates.

$$\delta b_{56}=\delta b_{65}=2(K_2+K_3)Sc_z, \tag{82d}$$

where $c\equiv\cos\psi$ and $s\equiv\sin\psi$. As we will see later, this interaction can only contribute significantly to the lowest-energy in-plane mode, where its effect is through the combination

$$\delta(a_{55}-a_{56}+b_{55}-b_{56})=4S(K_1-K_2c^2-K_3s^2)(1+c_z)$$
$$\equiv 4\Delta KS(1+c_z). \tag{83}$$

Note that $\Delta K=0$ for isotropic exchange.

A closely related interaction is the long-range dipolar interaction, whose contributions to the dynamical matrix are also evaluated in Appendix F. This interaction is dominant in $Sr_2CuO_2Cl_2$.[16] To include dipolar interactions we obtain (in Appendix F) the result

$$\delta(a_{55}-a_{56}+b_{55}-b_{56})=6g^2\mu_B^2S(1+c_z)X, \tag{84}$$

where X is the lattice sum

$$X=\sum_{j\in II:z_{ij}=c/2}\frac{x_{ij}y_{iy}\sigma_j}{r_{ij}^5}, \tag{85}$$

where i labels a fixed Cu_{II} site, σ_j is $+1$ if spins i and j are parallel and is -1 if they are antiparallel. Numerical evaluation yields

$$X=7\times10^{-4}\ \text{Å}^{-3}. \tag{86}$$

Therefore we should replace ΔK by

$$\Delta K_{\rm eff}=\Delta K+\tfrac{3}{2}g^2\mu_B^2X. \tag{87}$$

5. Cu_I-Cu_{II} interplanar interactions

Here we briefly summarize the results for a similar treatment of the Cu_I-Cu_{II} anisotropic interactions. The situation we consider is shown in the left panel of Fig. 5, where one sees that the isotropic component of the Cu_I-Cu_{II} interplanar interaction is frustrated. To describe the anisotropy of this interaction we introduce the principal axes for the Cu_I-Cu_{II} pair $\underset{\sim}{a}$-$\underset{\sim}{e}$, shown in the right panel of Fig. 5, as follows:

$$\hat m_1=-\hat y,\quad \hat m_2=\hat z\cos\phi-\hat x\sin\phi,$$
$$\hat m_3=-\hat z\sin\phi-\hat x\cos\phi. \tag{88}$$

The angle ϕ is not fixed by symmetry. We then write the anisotropic Cu_I-Cu_{II} interaction $\mathcal{H}_{ij}^{\text{I-II}}$ between nearest-neighboring spins i and j in adjacent planes as

$$\mathcal{H}_{ij}^{\text{I-II}}=\sum_{k=1}^{3}K_k'[\mathbf{S}_i\cdot\hat m_k(ij)][\mathbf{S}_j\cdot\hat m_k(ij)], \tag{89}$$

where $\hat m_k(ij)$ is the kth principal axes for the pair ij which can by obtained from the right panel of Fig. 5, by a rotation of coordinates, if necessary, and K_k' is associated principal value of the exchange tensor. In Appendix G we obtained the following contributions to the dynamical matrices for $q_x=q_y=0$:

$$\delta a_{15}=\delta b_{16}=\tfrac{1}{2}[K_1'+K_2'(1-3c^2)+K_3'(1-3s^2)]\equiv G_{\text{I-II}} \tag{90a}$$

$$\delta a_{16}=\delta b_{15}=\tfrac{1}{2}[K_1'+K_2'(1+c^2)+K_3'(1+s^2)]\equiv H_{\text{I-II}}, \tag{90b}$$

where $c\equiv\cos\phi$ and $s\equiv\sin\phi$. We will see later that these terms have a negligible effect on the spin-wave spectrum.

VI. SPIN-WAVE SPECTRUM

Explicitly, the dynamical matrices corresponding to the effective quadratic Hamiltonian containing the abovementioned anisotropies are of the form of Eq. (25) with

$$a_{11}=4JS+2J_3S+16C_2\tau+\zeta S+\alpha,$$

$$a_{12}=\Delta J_1S-4(6C_2-C_{2c}-4C_{2b})\tau,$$

$$a_{15}=\tfrac{1}{2}\Delta J_{12}S-\tfrac{1}{4}\zeta S+G_{\text{I-II}},$$

$$a_{16}=J_{12}^{(12)}S-\tfrac{1}{2}\Delta J_{12}S+\tfrac{1}{4}\zeta S+H_{\text{I-II}},$$

$$a_{55}=4J_2S-16\xi(2C_2-C_{2b})+2\alpha+4(K_1-K_2c^2-K_3s^2)S$$
$$+2(K_1+K_3c^2+K_2s^2)Sc_z, \tag{91}$$

$$a_{56}=2\Delta J_2S+16\xi(2C_2-3C_{2b})-\tfrac{3}{4}\zeta S$$
$$+2(K_2-K_3)(c^2-s^2)Sc_z,$$

$$b_{11}=8C_{2c}\tau,$$

$$b_{12}=2JS-\Delta J_1S-16C_{2b}\tau,$$

$$b_{15}=J_{12}^{(34)}S-\tfrac{1}{2}\Delta J_{12}S-\tfrac{1}{4}\zeta S+H_{\text{I-II}},$$

$$b_{16}=\tfrac{1}{2}\Delta J_{12}S+\tfrac{1}{4}\zeta S+G_{\text{I-II}},$$

TABLE I. Definitions of parameters. Notation: $\delta J \equiv (J_\perp - J_\parallel)/2$.

α	$C_\alpha \approx 0.1686$	τ	ζ	ξ	$C_2 \approx 0.01$ [a]	$J_{12}^{(n)}$
$C_\alpha J_{12}^2/J$	Eq. (48)	$(\delta J_1)^2/J$	$(\delta J_{12})^2/J_2$	$(\delta J_2)^2/J_2$	Eq. (C19)	Eqs. (43),(46)
ΔK	ΔK_{eff}	$X=7\times10^{-4}$ Å $^{-3}$				
Eq. (83)	Eq. (87)	Eq. (85)				

[a]See Ref. 13.

$$b_{55}=\tfrac{1}{4}\zeta S-16\xi C_{2b}+2(K_1-K_2s^2-K_3c^2)Sc_z, \quad (92)$$

$$b_{56}=4J_2S-2\Delta J_2S-\zeta S+48\xi C_{2b}+2(K_2+K_3)Sc_z. \quad (93)$$

[In the above tabulation we not have included dipolar interactions. These are easiest to include when we give the mode energies because these terms can then be combined via Eq. (87) with the pseudodipolar terms which we treated explicitly.] In Table I we summarize the definitions of the various parameters and in Table II we give estimates of their numerical values.

A. Cu_{II}'s ordered

1. Without 1/S renormalizations

Here we evaluate the energies of the four low-frequency modes in the presence of Cu_{II} ordering without any $1/S$ renormalizations. In what follows we will work to an accuracy of about 1%. That is, the only corrections of relative order $1/J$ we will keep are those of order J_{12}/J or J_2/J. Then, in the notation of Eqs. (34) and (37) the components of the large matrix $[\mathbf{A}+\sigma\mathbf{B}]_\sigma$ are

$$U_\sigma=8JS, \quad V_\sigma=2\sqrt{2}\sigma J_{12}S, \quad W_\sigma=8J_2S. \quad (94)$$

We neglect terms which are small compared to α and obtain

$$[\mathbf{A}-\mathbf{B}]_{\sigma=+1}=\begin{bmatrix} 4\Delta J_1S+\alpha+x_3 & -\sqrt{2}\alpha \\ -\sqrt{2}\alpha & 4\Delta J_2S+2\alpha \end{bmatrix} \quad (95)$$

for the out-of-plane sector, where $x_3=2J_3S(1-c_z)$, and

TABLE II. Estimated values of parameters from experiment and theory.

Parameter	Values in meV			
	From experiment		From theory	
	Value	Reference [a]	Value	Reference [a]
J	130 ± 5	7	145	28
J_3	0.14 ± 0.02	5,7		
J_{12}	-10 ± 2	7, TW		
J_2	10.5 ± 0.5	5		
$\Delta J_1(T=0$ K)	0.081 ± 0.01	TW	0.04	13,14
$\Delta J_1(T=200$ K)	0.068 ± 0.011	7		
ΔJ_{12}			1.3 [d]	27
ΔJ_2	0.004 ± 0.004	16, TW	0.036	26
δJ_1	±0.04	16, TW	-0.02	14
δJ_{12}	±0.027	4	-0.015	[b]
δJ_2			0.4	
ΔK_{eff}			2.73×10^{-4}	TW, Eq. (87)
α	0.135	5,7, TW	0.13	TW, App. B
τ	1.2×10^{-5}	16, TW	$\sim10^{-5}$	[c]
ζ	7×10^{-5}	4	2.2×10^{-5}	[c]
ξ			10^{-6}	[c]

[a]TW denotes this work.
[b]This is the contribution to δJ_{12} from dipolar interactions, which is much larger than that estimated from $\delta J/J \sim 1.5\times10^{-4}$.
[c]Evaluation based on the relevant J's.
[d]Evaluated for the similar compound $Ba_2Cu_3O_4Cl_2$.

$$[\mathbf{A}+\mathbf{B}]_{\sigma=-1}=\begin{bmatrix} \zeta S+\alpha+64C_2\tau+x_3 & \sqrt{2}(\alpha-\zeta S) \\ \sqrt{2}(\alpha-\zeta S) & 2(\zeta S+\alpha)-64\xi C_2+4\Delta K_{\rm eff}S(1+c_z) \end{bmatrix} \tag{96}$$

for the in-plane sector, where $\Delta K_{\rm eff}$ was defined by Eq. (87).

From Eq. (36) we get the higher frequency modes as

$$(\omega_+^{>})^2=8JS(4\Delta J_1 S+\alpha+x_3)-8J_{12}S\alpha+\frac{16J_2 S\alpha^2}{\alpha+4\Delta J_1 S+x_3}, \tag{97a}$$

$$(\omega_-^{>})^2=8JS(\alpha+x_3)+8S\alpha(-J_{12}+2J_2)-\frac{16J_2 S\alpha x_3}{\alpha+x_3}, \tag{97b}$$

and the lower frequency modes as

$$(\omega_+^{<})^2=64JJ_2S^2([4\Delta J_1 S+\alpha+x_3]\times[4\Delta J_2 S+2\alpha]-2\alpha^2)/(\omega_+^{>})^2, \tag{97c}$$

$$\begin{aligned}(\omega_-^{<})^2&=64JJ_2S^2([\zeta S+\alpha+64C_2\tau+x_3]\\&\quad\times[2\zeta S+2\alpha-64\xi C_2+4\Delta K_{\rm eff}S(1+c_z)]\\&\quad-2(\alpha-\zeta S)^2)/(\omega_-^{>})^2\\&\approx\left(\frac{64JJ_2S^2\alpha}{(\omega_-^{>})^2}\right)[2x_3+64(2\tau-\xi)C_2+8\zeta S\\&\quad+4\Delta K_{\rm eff}S(1+c_z)].\end{aligned} \tag{97d}$$

In obtaining the above results we replaced $UW-V^2$ by UW with an error of order 1%. To obtain the last line of Eq. (97d) we assumed that α dominates the other perturbations.

As we have already seen, quantum fluctuations of the frustrated Cu_I-Cu_{II} interactions cause $\omega_\sigma^{>}$ to be nonzero even if the exchange interactions are isotropic. When we introduce easy plane anisotropy (by making ΔJ_1 and/or ΔJ_2 nonzero) we introduce a gap into $\omega_+^{<}$, but $\omega_-^{<}$ has no gap yet, because without in-plane anisotropy a global rotation of spins within the easy plane costs no energy. The lowest mode develops a gap when we introduce the in-plane anisotropy and take account of quantum fluctuations. One might imagine that the strongest such anisotropy, namely that in J (scaled by the parameter δJ_1) would dominate in $\omega_-^{<}$. This effect is incorporated in the term proportional to $\tau=\delta J_1^2/J$, and indeed when the Cu_{II}'s are *not* ordered this term is the only one which contributes at $q_z=0$. However, when the Cu_{II}'s are ordered, the situation is different. Notice that this factor has no factor of S and more importantly, it is accompanied by the small numerical factor $C_2\approx0.01$. These observations remind us that this effect is another fluctuation effect. Within harmonic theory or mean-field theory the anisotropy of these Cu_I-Cu_I in-plane interactions averages to zero. In contrast, the weaker in-plane interaction between Cu_I's and Cu_{II}'s [scaled by $\zeta\equiv(\delta J_{12})^2/J$] appears already in mean-field theory.[4] Thus, this term, which is proportional to S, has no factor analogous to C_2 and it would dominate the term proportional to τ except for the fact (see next section) that its renormalization factor Z_ζ is quite small. However, when the Cu_{II}'s are ordered, the interplanar Cu_{II}-Cu_{II} dipolar interactions contained in $\Delta K_{\rm eff}$ are dominant, and lead to the dramatic increase in the effective fourfold anisotropy observed at low temperatures. The isotropic interplanar nearest-neighbor Cu_I-Cu_{II} are frustrated. The anisotropic Cu_I-Cu_{II} interlayer interactions (as embodied by the constants G and H) have only a negligible effect on the mode energies.

2. 1/*S* renormalizations

In this subsection we summarize how we incorporate the various renormalizations due to spin-wave interactions. We believe that the correct procedure is to calculate the mode energies correctly at first order in $1/S$ and then set $S=1/2$. Following this prescription we thereby obtain the following results:

$$(\omega_+^{>})^2=8JS[\alpha+4\Delta J_1 SZ_g^2+x_3Z_3^2]-8J_{12}S\alpha+\frac{16J_2S\alpha^2}{\alpha+4\Delta J_1SZ_g^2+x_3Z_3^2}, \tag{98a}$$

$$(\omega_-^{>})^2=8JS[\alpha+x_3Z_3^2]-8J_{12}S\alpha+\frac{16J_2S\alpha^2}{\alpha+x_3Z_3^2}, \tag{98b}$$

$$(\omega_+^{<})^2=64JJ_2S^2([4\Delta J_1SZ_g^2+\alpha+x_3Z_3^2]\times[4\Delta J_2SZ_g^2+2\alpha]-2\alpha^2)/(\omega_+^{>})^2, \tag{98c}$$

$$\begin{aligned}(\omega_-^{<})^2&=64JJ_2S^2([\zeta S+\alpha+64C_2\tau+Z_3^2x_3]\\&\quad\times[2\zeta S+2\alpha-64\xi C_2+4\Delta K_{\rm eff}S(1+c_z)]\\&\quad-2[\alpha-\zeta S]^2)/(\omega_-^{>})^2\\&\approx\left(\frac{64JJ_2S^2\alpha}{(\omega_-^{>})^2}\right)[2Z_3^2x_3+64(2\tau-\xi)C_2+8\zeta SZ_\zeta\\&\quad+4\Delta K_{\rm eff}S(1+c_z)Z_3^2].\end{aligned} \tag{98d}$$

Here we noted that spins not in the same plane are essentially uncorrelated and hence we have

$$J_3\to\tilde{Z}_3J_3,\quad \Delta K_{\rm eff}\to\tilde{Z}_3\Delta K_{\rm eff}, \tag{99}$$

where Eq. (40) gives $\tilde{Z}_3\approx1-0.2/S\to0.6$. But since J_3 and $\Delta K_{\rm eff}$ always enter the mode energies in combination with an isotropic exchange constant, we associate with them the renormalizations

$$J_3\to Z_3^2J_3,\quad \Delta K_{\rm eff}\to Z_3^2\Delta K_{\rm eff}, \tag{100}$$

TABLE III. Renormalizations $J \to ZJ$.

Quantity	J	J_3 [a]	$\sqrt{J\Delta J_I}$	ΔK_{eff} [b]
Renormalized to	$Z_c J$	$\tilde{Z}_3 J_3$	$Z_g \sqrt{J\Delta J_I}$	$\Delta K_{\mathrm{eff}} \tilde{Z}_3^2$
	$(1+0.085/S)J$	$(1-0.2/S)J_3$	$(1-0.2/S)\sqrt{J\Delta J_I}$	$(1-0.2/S)K_{\mathrm{eff}}$
Refer to	Ref. 21	Eq. (40)	Ref. 25	Eq. (99)

[a]In the dynamics $JJ_3 \to Z_c \tilde{Z}_3 JJ_3 \equiv Z_3^2 JJ_3$, where we set $Z_3^2 = 0.77$.

[b]In the dynamics $J_2 \Delta K_{\mathrm{eff}} \to Z_c \tilde{Z}_3 J_2 \Delta K_{\mathrm{eff}} \equiv Z_3^2 J_2 \Delta K_{\mathrm{eff}}$, where we set $Z_3^2 = 0.77$.

where $Z_3^2 = \tilde{Z}_3 Z_c$. Thus $Z_3^2 = (1-0.2/S)(1+0.085/S) = (1-0.115/S) \to 0.77$. Also, we will determine Z_ζ by comparison, in Eq. (108) below, with the phenomenological treatment[4] of the statics. For convenience we summarize in Table III the renormalizations of the various interactions which follow from our treatment to order $1/S$.

B. $\mathrm{Cu_{II}}$'s disordered

To get the energies of the spin-wave modes when the $\mathrm{Cu_{II}}$'s are disordered one sets $J_{12} = J_2 = 0$ (i.e., modes $\omega_+^<$ and $\omega_-^<$ no longer exist as elementary excitations) and $\alpha = 0$, in which case we get

$$(\omega_+)^2 = 8JS[4\Delta J_1 S Z_g^2 + 2J_3 S Z_3^2 (1-c_z)], \quad (101a)$$

$$(\omega_-)^2 = 8JS[64\tau C_2 + 2J_3 S Z_3^2 (1-c_z)]. \quad (101b)$$

Note that in Eq. (97b) we had dropped a term representing the fourfold anisotropy which is proportional to τ, because such a term is negligible in comparison to α. Here, with α not present, we restore this term in ω_-. Note also that the higher energy mode is the one which has fluctuations out of the plane (as indicated by the dependence on ΔJ_1) and at zero wave vector is of the expected form $\omega^2 = 2H_E H_A$, with the exchange field $H_E = 4JS$ and the anisotropy field $H_A = 4\Delta J_1 S$. The energy of this out-of-plane gap is about 5 meV in many lamellar copper oxide antiferromagnets.[2] The lower-energy mode involves motion of the spins within the plane and would have no gap at zero wave vector except for the appearance of a small effective fourfold anisotropy, which was obtained previously[13] from phenomenological considerations. The same result for the gap, namely, $\omega = 16\delta J_1 \sqrt{2C_2 S} \approx 1.6\delta J_1$, is obtained from the microscopic calculation given in Appendix C and also in Ref. 25.

C. Comparison of static and dynamic theories

Here we briefly compare our results with those of a mean-field treatment of the statics.[4] In that calculation the fourfold anisotropy is included phenomenologically and the anisotropic $\mathrm{Cu_I}$-$\mathrm{Cu_{II}}$ interactions are included even when the $\mathrm{Cu_{II}}$ sublattice is not antiferromagnetically ordered. When the $\mathrm{Cu_{II}}$ sublattice is ordered, the static treatment assumes that the Shender mechanism is strong enough that all spins are essentially collinear. So the dynamics of the Goldstone mode should involve the static response coefficients, although spin-wave hydrodynamics[29] rigorously applies only in the limit of zero frequency.

Since the statics treat the fourfold anisotropy phenomenologically, as did Yildirim *et al.*,[13] we identify their fourfold anisotropy constant K, which scales the anisotropy energy per $\mathrm{Cu_I}$ spin, from

$$E = -\tfrac{1}{2} K \cos(4\theta), \quad (102)$$

because there are two $\mathrm{Cu_I}$'s per unit cell. Also θ is the angle of the magnetic moment with respect to the easy (1,0,0) axis. In Ref. 13 the energy per $\mathrm{Cu_I}$ spin is (in the present notation)

$$E = 32 C_2 \tau S (S_x^2 S_y^2 / S^4). \quad (103)$$

So we make the identification $K = 8C_2 \tau S$, or, if we include the effects of the $\mathrm{Cu_{II}}$'s,

$$K = 4C_2(2\tau - \xi)S. \quad (104)$$

We start by comparing the results of the two approaches when the $\mathrm{Cu_{II}}$'s are disordered. There the spin-wave calculation completely ignores the presence of the $\mathrm{Cu_{II}}$'s, whereas in the statics the $\mathrm{Cu_{II}}$'s are characterized by their susceptibility in the pseudodipolar field caused by the small in-plane anisotropy of the $\mathrm{Cu_I}$-$\mathrm{Cu_{II}}$ interactions. In the statics for temperatures far below the ordering temperature for the $\mathrm{Cu_I}$ sublattice (but still with the $\mathrm{Cu_{II}}$'s disordered) one has the effective fourth-order anisotropy constant k_{stat} from the statics as

$$k_{\mathrm{stat}} = 2K + 8M_0^2 J_{12}^2 \chi_{\mathrm{I}} [1 - 8\chi_{\mathrm{II}}^2 J_{12}^2]^{-1}, \quad (105)$$

where we introduce the Cu spin susceptibilities $\chi_{\mathrm{I}} \approx 0.53/(8J)$, $\chi_{\mathrm{II}} \approx 0.53/(8J_2)$, and (in the present notation)

$$M_0 = 4\delta J_{12} \langle S \rangle \chi_{\mathrm{II}}, \quad (106)$$

where $\langle \cdots \rangle$ denotes a thermal average. If one takes $\delta J_{12} = 0.025$ meV, then $M_0 = 2 \times 10^{-4}$. Then the second term on the right-hand side of Eq. (105) is about 6×10^{-8} meV, compared with $2K$ which was found[4] to be 2×10^{-6} meV. So this correction (due to paramagnetic $\mathrm{Cu_{II}}$'s) which is absent from our spin-wave analysis is negligible.

When the $\mathrm{Cu_{II}}$'s are well ordered, Ref. 4 gives approximately

$$k_{\mathrm{stat}} = 2K + 8(\delta J_{12})^2 \langle S \rangle^2 [0.53/(8J_2)]. \quad (107)$$

Using Eq. (104) as the identification of K, we see from Eq. (98d) that the mode energy involves the combination (for $\xi \ll \tau$ and $\zeta S \ll \alpha$) which we identify to be the effective value of k from the dynamics, k_{dyn}, where

TABLE IV. Experimental values of spin-wave gaps at zero wave vector.

Mode	Temperature	Energy (meV)	Ref.
$\omega_+^>$	$T=200$ K	5.5(3) [a]	5
$\omega_-^>$	$T=200$ K	0.066(4)	16
$\omega_+^>$	$T\to 0$ K	10.8(6)	7
$\omega_-^>$	$T\to 0$ K	9.1(3)	7
$\omega_+^<$	$T\to 0$ K	1.7473(4)	16
$\omega_+^<$	$T\to 0$ K	1.72(20)	7
$\omega_-^<$	$T\to 0$ K	0.149(3)	16

[a]Extrapolated to $T=0$.

$$k_{\rm dyn}=8(2\tau-\xi)C_2S+\zeta S^2Z_\zeta+\Delta K_{\rm eff}S^2Z_3^2$$
$$=2K+(\delta J_{12})^2S^2Z_\zeta/J_2+\Delta K_{\rm eff}S^2Z_3^2. \quad (108)$$

We see that the term $(0.53)\langle S\rangle^2$ in the statics appears as S^2Z_ζ in the spin-wave dynamics. With an appropriate renormalization $Z_\zeta\approx 0.19$, these two terms are the same. Thus, as far as the intralayer interactions are concerned the comparison between statics and dynamics indicates that these terms are correctly treated. We also see that the treatment of the statics did not include the interplanar anisotropic interaction, $\Delta K_{\rm eff}$. As we shall see, this term gives an important contribution to the mode $\omega_-^<$, so it should be included in a reanalysis of the statics. In terms of the constant $k_{\rm dyn}$ we may write Eq. (98d) as

$$(\omega_-^<)^2(\mathbf{q}=0)=64J_2k_{\rm dyn}\left(\frac{J}{J-J_{12}+2J_2}\right). \quad (109)$$

Thus we conclude that except for the fact that the statics ignored the interplanar anisotropic Cu_{II}-Cu_{II} interactions, the two theoretical approaches are compatible with one another. In the next section will show that the *experimental* results from static and dynamic measurements are also consistent with one another.

D. Comparison to experiments

The comparison between the present theory and experiments has been described briefly in several previous publications.[3,5,16] Since a more detailed comparison is given in paper I, we will simply summarize the comparison of the theoretical and experimental results. First one has the estimate for J which is nearly the same for all cuprates. This estimate has been refined by Kim,[30–32] who gives $J=130$ meV. The value $J_2=10.5$ meV has been accurately determined[5] by comparing the experimental dispersion with respect to in-plane wave vector of Cu_{II} spin waves to various theoretical treatments which take account of spin-wave interactions.[33]

Now we discuss the analysis of the magnon gaps at zero wave vector where values are listed in Table IV. We first fit the observed[16] in-plane gap when the Cu_{II}'s are disordered. Equation (101b) yields $\omega_-=16\sqrt{2C_2S}\,\delta J_1$ and with[16] $\omega_-=0.066$ meV, we get $|\delta J_1|=|J_\parallel-J_\perp|/2=0.042$ meV, a value which is about twice the theoretical estimates.[14] Using Eqs. (104) and (105) this corresponds to $k=16C_2S\tau=16(0.01)(0.5)(0.042)^2/130=1\times10^{-6}$ meV, compared to the value deduced from the statics[4] $k=2\times10^{-6}$ meV, for 70 K$\leqslant T\leqslant$120 K. At low temperature ($T=1.4$ K), where the Cu_{II}'s are well ordered, the statics[34] gives $k=25\times10^{-6}$ meV. From Eq. (109) with $\omega_-^<=0.15$ meV, we get $k_{\rm dyn}=41\times10^{-6}$ meV. These results are listed in Table V, where we see only a qualitative consistency between the interpretation of the static and dynamic experiments. It is possible that the quantum renormalizations (which affect the determination of k from the observed mode energy) are not quite correct. Also, the interpretation of the statics within which the Cu_{II}-Cu_{II} interplanar anisotropy is subsumed into the fourfold anisotropy constant k is not strictly correct. If we fix δJ_1 to fit the value of ω_- at $T=100$ K and assume that the interplanar Cu_{II}-Cu_{II} interactions result from the actual dipole-dipole interactions, then the temperature dependence of k results from the last term in Eq. (108). With only dipolar (i.e., no pseudodipolar) interactions, Eqs. (87) and (86) give (with $g=2.2$[4]) $\Delta K_{\rm eff}=273\times10^{-6}$ meV, so that $\Delta K_{\rm eff}S^2Z_3^2=53\times10^{-6}$ meV, from which $k_{\rm dyn}=56\times10^{-6}$ meV. From Table V it is clear that the experimentally deduced temperature dependence of k is qualitatively accounted for by the intraplanar dipolar interactions.

Now we consider the higher-energy modes. Fitting to the observed[7] energy $\omega_+=5.5$ meV of the out-of-plane gap when the Cu_{II}'s are disordered to Eq. (101a) (with $Z_g=0.6$) we obtain the value of $\Delta J_1=0.081$ meV. As was the case for δJ_1, this result is also about twice the theoretical estimates for a simple CuO plane.[13,14] Given the values of these parameters, both higher-energy modes at low temperature involve only the one additional parameter α. If we determine α from $\omega_+^>$ we get $\alpha=0.14$ meV, whereas if we determine α from $\omega_-^>$ we get $\alpha=0.13$ meV. These two values agree perfectly with one another and their average coincides with the theoretical evaluation of Appendix B that $\alpha=0.13$ meV. Clearly these agreements strongly support our interpretation of the role of fluctuations embodied by the

TABLE V. Values in (10^{-6} meV) of the fourfold anisotropy constant k.

k	$T=1.4$ K	$T=100$ K
Experimental: From statics (Ref. 4)	25	2
Experimental: Fitting Eq. (109) to AFMR data (Ref. 16)	41	1
Theoretical: See Eq. (108)	56	1 [a]

[a]$|J_\perp-J_\parallel|=0.041$ meV is fixed so that the dynamics and theory agree.

parameter α. Note that a biquadratic interaction between two spin 1/2's can be subsumed into a ordinary Heisenberg exchange interaction. Therefore biquadratic exchange cannot contribute to α.

Finally we consider the lower-energy out-of-plane mode in the zero temperature limit. The AFMR data[15,16] gives $\omega_+^< = 1.7473(4)$ meV, more accurate than, but entirely consistent with, the data of Ref. 7. Evaluating the expression in Eq. (98c) with $\Delta J_2 = 0$ gives $\omega_+^< = 1.717$ meV. If we fix ΔJ_2 to fit the experimental value of this gap, we get $\Delta J_2 = 0.004 \pm 0.004$ meV. We attribute a large uncertainty to ΔJ_2 because its value changes significantly if ΔJ_1 or α is slightly modified. To get the same relative out-of-plane anisotropy, $\Delta J/J$, for the Cu_{II}-Cu_{II} exchange as for the Cu_I-Cu_I exchange would require $\Delta J_1 = 0.008$ meV.

VII. DYNAMIC STRUCTURE FACTOR

The cross section, $\sigma(\mathbf{q},\omega)$, for inelastic neutron scattering from magnetic ions is proportional to the dynamic structure factor $S^{\alpha\beta}(\mathbf{q},\omega)$ which in turn is related to the spin-spin correlation function. We have

$$\sigma(\mathbf{q},\omega) \propto \sum_{\alpha\beta} (\delta_{\alpha,\beta} - \hat{q}_\alpha \hat{q}_\beta) S^{\alpha\beta}(\mathbf{q},\omega). \tag{110}$$

According to the fluctuation-dissipation theorem, we may write

$$S^{\alpha\beta}(\mathbf{q},\omega) = \frac{1}{\pi} n(\omega) \mathrm{Im}\, \chi^{\alpha\beta}(\mathbf{q},\omega - i0^+), \tag{111}$$

where $n(\omega) = [e^{\hbar\omega/(kT)} - 1]^{-1}$ and, in the usual notation,[35] the **A**-**B** Green's function is defined as

$$\langle\langle \mathbf{A};\mathbf{B} \rangle\rangle_\omega = \sum_{m,n} p_n \left[\frac{\langle n|\mathbf{A}|m\rangle\langle m|\mathbf{B}|n\rangle}{\omega - E_m + E_n} - \frac{\langle n|\mathbf{B}|m\rangle\langle m|\mathbf{A}|n\rangle}{\omega + E_m - E_n} \right], \tag{112}$$

where $|n\rangle$ and $|m\rangle$ are exact eigenstates with respective energies E_n and E_m and p_n is the Boltzmann weight of the state $|n\rangle$. Then χ, the dynamic susceptibility, is written as the Green's function

$$\chi^{\alpha,\beta}(\mathbf{q},\omega) = \langle\langle S^\alpha(\mathbf{q}); S^\beta(-\mathbf{q}) \rangle\rangle_\omega. \tag{113}$$

We construct the dynamic susceptibility by writing the spin operators in terms of boson operators at leading order in $1/S$:

$$\begin{aligned} S^+(\mathbf{q}) &= \sqrt{2S}[a(\mathbf{q}) + b^\dagger(-\mathbf{q}) + c^\dagger(-\mathbf{q}) \\ &\quad + d(\mathbf{q}) + e^\dagger(-\mathbf{q}) + f(\mathbf{q})], \\ S^-(-\mathbf{q}) &= \sqrt{2S}[a^\dagger(\mathbf{q}) + b(-\mathbf{q}) + c(-\mathbf{q}) \\ &\quad + d^\dagger(\mathbf{q}) + e(-\mathbf{q}) + f^\dagger(\mathbf{q})]. \end{aligned} \tag{114}$$

Thus we have

$$\begin{aligned} S^\eta(\mathbf{q}) &= [S^+(\mathbf{q}) + S^-(\mathbf{q})]/2 \\ &= \sqrt{S/2} \sum_m [V_m(\eta)\xi_m(\mathbf{q}) + V_m(\eta)^* \xi_m^\dagger(-\mathbf{q})] \end{aligned} \tag{115}$$

and

$$\begin{aligned} S^z(\mathbf{q}) &= -i[S^+(\mathbf{q}) - S^-(\mathbf{q})]/2 \\ &= \sqrt{S/2} \sum_m [V_m(z)\xi_m(\mathbf{q}) + V_m(z)^* \xi_m^\dagger(-\mathbf{q})], \end{aligned} \tag{116}$$

where the operators are labeled as in Eq. (15) and the transpose of the column vectors $\mathbf{V}(\alpha)$ is

$$\tilde{V}(\eta) = (1,1,1,1,1,1), \quad \tilde{V}(z) = i(1,-1,-1,1,-1,1). \tag{117}$$

Thus we may write

$$\begin{aligned} \chi^{\alpha\beta}(\mathbf{q},\omega) = \tfrac{1}{2} S \sum_{mn} \langle\langle [V_m(\alpha)\xi_m(\mathbf{q}) + V_m(\alpha)^* \xi_m^\dagger(-\mathbf{q})]; \\ [V_n(\beta)\xi_n(-\mathbf{q}) + V_n(\beta)^* \xi_n^\dagger(\mathbf{q})] \rangle\rangle_\omega. \end{aligned} \tag{118}$$

We may evaluate these response functions in terms of normal modes. Suppose we have found the unnormalized right eigenvectors of the dynamical matrix, Eq. (23). That is we have the column vectors Φ_j which satisfy

$$[\mathbf{A}+\mathbf{B}][\mathbf{A}-\mathbf{B}]\Phi_j = \omega_j^2 \Phi_j. \tag{119}$$

Then we make the identification that

$$\mathbf{P}_j - \mathbf{Q}_j = x_j \Phi_j. \tag{120}$$

We can arbitrarily fix the phase of the normal mode operators so that x_j is real positive. Then

$$[\mathbf{A}-\mathbf{B}]x_j\Phi_j = [\mathbf{A}-\mathbf{B}][\mathbf{P}_j - \mathbf{Q}_j] = \omega_j[\mathbf{P}_j + \mathbf{Q}_j] \tag{121}$$

or

$$\mathbf{P}_j + \mathbf{Q}_j = (x_j/\omega_j)[\mathbf{A}-\mathbf{B}]\Phi_j, \tag{122}$$

so that

$$\begin{aligned} \mathbf{P}_j &= \frac{x_j}{2}(\mathcal{I} + \omega_j^{-1}[\mathbf{A}-\mathbf{B}])\Phi_j \\ \mathbf{Q}_j &= \frac{x_j}{2}(-\mathcal{I} + \omega_j^{-1}[\mathbf{A}-\mathbf{B}])\Phi_j. \end{aligned} \tag{123}$$

To use Eq. (18) we write

$$\mathbf{P}_j^\dagger \mathbf{P}_j - \mathbf{Q}_j^\dagger \mathbf{Q}_j = \frac{x_j^2}{\omega_j}(\Phi_j^\dagger [\mathbf{A}-\mathbf{B}]\Phi_j), \tag{124}$$

so that

$$x_j^2 = \frac{\omega_j}{(\Phi_j^\dagger[\mathbf{A}-\mathbf{B}]\Phi_j)}. \tag{125}$$

Then we write the susceptibilities as

$$\begin{aligned}(2/S)\chi^{\alpha\beta}(\mathbf{q},\omega) &= \sum_{m,n,r}[V_m(\alpha)P_{mr}(\mathbf{q})+V_m(\alpha)^*Q_{mr}(\mathbf{q})][V_n(\beta)^*P_{nr}(\mathbf{q})^*+V_n(\beta)Q_{nr}(\mathbf{q})^*]\langle\langle\tau_r(\mathbf{q});\tau_r^\dagger(\mathbf{q})\rangle\rangle_\omega \\ &+\sum_{mnr}[V_m(\alpha)Q_{mr}(\mathbf{q})+V_m(\alpha)^*P_{mr}(\mathbf{q})][V_n(\beta)^*Q_{nr}(\mathbf{q})^*+V_n(\beta)P_{nr}(\mathbf{q})^*]\langle\langle\tau_r^\dagger(\mathbf{q});\tau_r(\mathbf{q})\rangle\rangle_\omega \\ &=\sum_r (\{[\tilde{\mathbf{V}}(\alpha)\mathbf{P}_r]+[\tilde{\mathbf{V}}(\alpha)^*\mathbf{Q}_r]\}\{[\tilde{\mathbf{V}}(\beta)\mathbf{P}_r]+[\tilde{\mathbf{V}}(\beta)^*\mathbf{Q}_r]\}^*[\omega-\omega_r(\mathbf{q})]^{-1}+\{[\tilde{\mathbf{V}}(\alpha)\mathbf{Q}_r]+[\tilde{\mathbf{V}}(\alpha)^*\mathbf{P}_r]\} \\ &\times\{[\tilde{\mathbf{V}}(\beta)\mathbf{Q}_r]+[\tilde{\mathbf{V}}(\beta)^*\mathbf{P}_r]\}^*[\omega+\omega_r(\mathbf{q})]^{-1}) \\ &\equiv\sum_r\left[\frac{J_r^{\alpha\beta}(\mathbf{q})}{\omega-\omega_r(\mathbf{q})}+\frac{I_r^{\alpha\beta}(\mathbf{q})}{\omega+\omega_r(\mathbf{q})}\right],\end{aligned} \tag{126}$$

where we left the argument **q** implicit in several places. We will refer to I and J as ''intensities,'' although to get inelastic neutron scattering cross-sections one needs to include several other factors. At low temperature we only need

$$\begin{aligned}I_r^{\alpha\beta}(\mathbf{q}) &= x_r^2(\delta_{\alpha,z}[V(z)^\dagger\Phi_r(\mathbf{q})]+\omega_r(\mathbf{q})^{-1}\delta_{\alpha,\eta} \\ &\times\{V(\eta)^\dagger[\mathbf{A}-\mathbf{B}]\Phi_r(\mathbf{q})\})(\delta_{\beta,z}[V(z)^\dagger\Phi_r(\mathbf{q})] \\ &+\omega_r(\mathbf{q})^{-1}\delta_{\beta,\eta}\{V(\eta)^\dagger[\mathbf{A}-\mathbf{B}]\Phi_r(\mathbf{q})\})^*.\end{aligned} \tag{127}$$

In writing this result we used the fact that $\mathbf{V}(\eta)$ is real and $\mathbf{V}(z)$ is imaginary. From now on, we specialize to the case of wave vectors of the form $\mathbf{q}=\mathbf{G}+q_z\hat{z}$. In that case $I_r^{\eta z}+I_r^{z\eta}$ vanishes and

$$I_r^{zz}=\frac{|[\mathbf{V}(z)^\dagger\Phi_r(\mathbf{q})]|^2\omega_r(\mathbf{q})}{\{\Phi_r(\mathbf{q})^\dagger[\mathbf{A}-\mathbf{B}]\Phi_r(\mathbf{q})\}} \tag{128a}$$

$$I_r^{\eta\eta}=\frac{|\{\mathbf{V}(\eta)^\dagger[\mathbf{A}-\mathbf{B}]\Phi_r(\mathbf{q})\}|^2}{\omega_r(\mathbf{q})\{\Phi_r(\mathbf{q})^\dagger[\mathbf{A}-\mathbf{B}]\Phi_r(\mathbf{q})\}}. \tag{128b}$$

The above results are useful for the out-of-plane $(\sigma=+1)$ modes in which case $[\mathbf{A}-\mathbf{B}]$ is the small matrix. Alternatively, for in-plane $(\sigma=-1)$ modes when $\mathbf{A}+\mathbf{B}$ is the small matrix the following forms are useful:

$$I_r^{zz}=\frac{|\{\mathbf{V}(z)^\dagger[\mathbf{A}+\mathbf{B}]\Psi_r(\mathbf{q})\}|^2}{\omega_r(\mathbf{q})\{\Psi_r(\mathbf{q})^\dagger[\mathbf{A}+\mathbf{B}]\Psi_r(\mathbf{q})\}} \tag{129a}$$

$$I_r^{\eta\eta}=\frac{|[\mathbf{V}(\eta)^\dagger\Psi_r(\mathbf{q})]|^2\omega_r(\mathbf{q})}{\{\Psi_r(\mathbf{q})^\dagger[\mathbf{A}+\mathbf{B}]\Psi_r(\mathbf{q})\}}. \tag{129b}$$

For high symmetry directions of the wave vector, the matrices **A** and **B** may be brought into block diagonal form by a unitary transformation **U**. In that case we may apply the above formulas in terms of the transformed quantities indicated by primes:

$$\mathbf{A}'\equiv\mathbf{U}^\dagger\mathbf{A}\mathbf{U},\quad \mathbf{B}'\equiv\mathbf{U}^\dagger\mathbf{B}\mathbf{U},$$

$$\Phi_r'\equiv\mathbf{U}^\dagger\Phi_r,\quad \Psi_r'\equiv\mathbf{U}^\dagger\Psi_r,\quad \mathbf{V}'(\alpha)\equiv\mathbf{U}^\dagger\mathbf{V}(\alpha). \tag{130}$$

For wave vectors which are equal modulo a reciprocal lattice vector, the corresponding quantities $\mathbf{A}'$, $\mathbf{B}'$, Φ', and Ψ' are equal. However, the intensities at such equivalent points will differ because **U**, and hence $\mathbf{V}'$, depend specifically on the zone of the wave vector. This can be seen explicitly in Appendix A where we obtain the results summarized in Tables VI and VII. Note that the $\sigma=+1$ sector does have intensity mainly in I^{zz} in confirmation of our identification of this as the out-of-plane sector. Similarly, the $\sigma=-1$ sector has its intensity mainly in $I^{\eta\eta}$ as expected for in-plane modes. These identifications are also consistent with the fact that the $\sigma=+1$ modes depend on the out-of-plane anisotropies scaled by the ΔJ's, whereas the $\sigma=-1$ modes do not involve these quantities.

VIII. CONCLUSIONS

Here we briefly summarize the significant conclusions from this work.

(1) The degeneracy, present within mean-field theory, in which the Cu_{II} sublattice spins can be globally rotated with respect to the Cu_I spins is removed by quantum fluctuations which cause the sublattice magnetizations to be collinear, as first indicated by Shender.[8]

(2) A degeneracy present within mean-field and linear spin-wave theories, in which the magnetization can be globally rotated through an arbitrary angle within the easy plane

TABLE VI. Amplitude of the dynamic structure factor. Results for wave vector $2\pi(H\hat{x}/a+K\hat{y}/a+L\hat{z}/c)+q_z$ for H and K half integral and $H+K+L$ an even integer. Results are given only to leading order in J. $y_3=x_3+4\Delta J_1S+\frac{1}{2}\alpha=2J_3S[1-\cos(q_zc/2)]+4\Delta J_1S+\frac{1}{2}\alpha$. The mode energies (without $1/S$ corrections) and intensities $[I_r^{\alpha\beta}(\mathbf{q})]$ are independent of the particular values of H, K, and L and are evaluated for $q_z=0$.

Mode energy	Energy [a] (meV)	Intensity Formula	Intensity Evaluation
$\omega_+^> = [8JSy_3]^{1/2}$	10.8	$I_{>+}^{zz}=0$	0
		$I_{>+}^{\eta\eta}=0$	0
$\omega_+^< = \left\{8J_2S\left[4\Delta J_2S+\frac{2\alpha(4\Delta J_1S+x_3)}{y_3}\right]\right\}^{1/2}$	1.72	$I_{<+}^{zz}=\frac{16J_2S}{\omega_+^<}$	12
		$I_{<+}^{\eta\eta}=0$	0
$\omega_-^> = [8JS(x_3+2\alpha)]^{1/2}$	9.1	$I_{>-}^{zz}=0$	0
		$I_{>-}^{\eta\eta}=0$	0
$\omega_-^< = \left[8J_2S\frac{2\alpha x_3+64\alpha(2\tau-\xi)C_2+8\alpha\zeta S}{x_3+\alpha}\right]^{1/2}$	0.15	$I_{<-}^{zz}=0$	0
		$I_{<-}^{\eta\eta}=\frac{16J_2S}{\omega_-^<}$	140

[a]See Table IV.

is similarly removed by quantum fluctuations, as first proposed in Ref. 13.

(3) These fluctuation effects, in addition to selecting the ground state from among the classically degenerate configurations, also give rise to nonzero energies of the corresponding spin-wave excitations. The most dramatic evidences of this phenomenon are the striking increases of the out-of-plane gap energy from 5 to 10 meV and that of the in-plane

TABLE VII. Amplitude of the dynamic structure factor. Results for wave vector $2\pi(H\hat{x}/a+K\hat{y}/a+L\hat{z}/c)+q_z$ for H and K integers and $H+K+L$ an even integer. The notation is as in Table V. Results are given only to leading order in J. The intensities are evaluated for $q_z=0$ and $H=L=1$ and $K=0$.

Mode energy [a] (meV)	Formula for intensity	Intensity
$\omega_+^> = 10.8$	$I_{>+}^{zz}=\frac{8JS}{\omega_+^>}[1-(-1)^L]^2$	50
	$I_{>+}^{\eta\eta}=\frac{8JS}{\omega_+^{>3}}\lvert y_3[1+(-1)^L]+(-1)^H\alpha\rvert^2$	0
$\omega_+^< = 1.72$	$I_{<+}^{zz}=[1-(-1)^L]^2\frac{(8JS)^2(4J_2S)\alpha^2}{\omega_+^{>4}\omega_+^<}$	26
	$I_{<+}^{\eta\eta}=\frac{\omega_+^<}{4J_2S}$	0
$\omega_-^> = 9.1$	$I_{>-}^{zz}=\frac{8JS}{\omega_-^{>3}}\{[1+(-1)^L][x_3+\frac{1}{2}\alpha]+\alpha(-1)^H\}^2$	0
	$I_{>-}^{\eta\eta}=\frac{8JS}{\omega_-^>}[1-(-1)^L]^2$	59
$\omega_-^< = 0.15$ meV	$I_{<-}^{zz}=\frac{\omega_-^<}{4J_2S}$	0
	$I_{<-}^{\eta\eta}=\frac{(4J_2S)(8JS)^2\alpha^2}{\omega_-^<\omega_-^{>4}}[1-(-1)^L]^2$	570

[a]See Table IV.

gap from zero to 9 meV when the Cu_{II} sublattice evolves from disorder to order.

(4) The experimental results of inelastic neutron scattering for the lowest energy gaps are broadly consistent with the effective fourfold anisotropy previously obtained from the statics experiments.[4] More precise agreement may depend on more accurate understanding of the various renormalizations due to quantum and thermal fluctuations.

(5) Our improved theoretical treatment which now includes the interlayer dipolar interactions resolves the mystery surrounding the dramatic increase (first found in the statics[4]) in the effective fourfold anisotropy as the temperature is reduced into the regime where the Cu_{II}'s order. In fact the dipolar interlayer interactions between the Cu_{II}'s dominates the effective fourfold anisotropy when the Cu_{II}'s develop long range order.

(6) Recent AFMR results[16] lead to an identification of the small in-plane anisotropies and qualitatively confirm previous theoretical estimates of the exchange anisotropy induced by spin-orbit interactions.[13,14]

ACKNOWLEDGMENTS

This work was supported by the U.S.-Israel Binational Science Foundation. R.J.B. and Y.J.K. were also supported by NSF Grant No. DMR97-04532 and by the MRSEC Program of the NSF under Grant No. DMR98-08941 (at MIT), under Contract No. DE-AC02-98CH10886, Division of Material Science, U.S. Department of Energy (at BNL), and by the NSF under Agreement No. DMR-9243101 (at NIST). We thank M. A. Kastner for useful discussions.

APPENDIX A: INTENSITY CALCULATIONS

In this appendix we evaluate the intensities for which formulas are given in Sec. V. We first give the unitary transformation which brings the matrices **A** and **B** into block diagonal form. We do this for wave vectors $\mathbf{q}=\mathbf{G}+q_z\hat{z}$, where

$$\mathbf{G}=2\pi\left[\frac{H\hat{x}}{a}+\frac{K\hat{y}}{a}+\frac{L\hat{z}}{c}\right], \tag{A1}$$

where H and K are either both half integral or both integral and $H+K+L$ is an even integer. Then

$$\mathbf{U}=\frac{1}{2}\begin{bmatrix} \sqrt{2} & 0 & 1 & 0 & 1 & 0 \\ 0 & \sqrt{2} & (-1)^{H+K} & 0 & -(-1)^{H+K} & 0 \\ 0 & -(-1)^{2H}\sqrt{2} & (-1)^{H-K} & 0 & -(-1)^{H-K} & 0 \\ -(-1)^{2H}\sqrt{2} & 0 & (-1)^{2H} & 0 & (-1)^{2H} & 0 \\ 0 & 0 & 0 & (-i)^{2H}\sqrt{2} & 0 & (-i)^{2H}\sqrt{2} \\ 0 & 0 & 0 & (i)^{2H}\sqrt{2} & 0 & -(i)^{2H}\sqrt{2} \end{bmatrix}. \tag{A2}$$

The first two columns are the high frequency Cu_I optical modes. Columns Nos. 3 and 4 are the $\sigma=1$ out-of-plane modes and columns Nos. 5 and 6 are the $\sigma=-1$ in-plane modes. The following results hold for all wave vectors of the form $\mathbf{q}=\mathbf{G}+q_z\hat{z}$.

1. Out-of-plane modes

For the out-of-plane sector we have (for dominant J)

$$\mathbf{A}'-\mathbf{B}'=\begin{pmatrix} x_3+4\Delta J_1S+\frac{1}{2}\alpha & \alpha/\sqrt{2} \\ \alpha/\sqrt{2} & 4\Delta J_2S+\alpha \end{pmatrix},$$

$$\mathbf{A}'+\mathbf{B}'=\begin{pmatrix} 8JS & \sqrt{2}J_{12}S \\ \sqrt{2}J_{12}S & 8J_2S \end{pmatrix} \tag{A3}$$

independent of **G**, where $x_3=2J_3S[1-\cos(cq_z/2)]$. Note that q_z is measured relative to the reciprocal lattice vector in question. We now tabulate the right eigenvectors of the block matrices $M_{+-}\equiv[\mathbf{A}'+\mathbf{B}'][\mathbf{A}'-\mathbf{B}']$ associated with the eigenvalues (the squares of the mode energies) ω_r^2. We have

$$\tilde{\Phi}_+^{>}=[1,0],\quad (\omega_+^{>})^2=(8JS)(x_3+4\Delta J_1S+\tfrac{1}{2}\alpha),$$

$$\tilde{\Phi}_+^{<}=[-\alpha/\sqrt{2},x_3+4\Delta J_1S+\tfrac{1}{2}\alpha],$$

$$(\omega_+^{<})^2=(8J_2S)\left[4\Delta J_2S+\alpha-\frac{\frac{1}{2}\alpha^2}{x_3+4\Delta J_1S+\frac{1}{2}\alpha}\right]. \tag{A4}$$

Also we find that

$$\mathbf{V}(z)'=\begin{bmatrix} 1-(-1)^{H+K} \\ 0 \end{bmatrix},$$

$$\mathbf{V}(\eta)' = \begin{bmatrix} 1+(-1)^{H+K} \\ \sqrt{2}(-1)^{H}, \end{bmatrix} \text{ for integer } H, \tag{A5a}$$

$$\mathbf{V}(z)' = \begin{bmatrix} 0 \\ i^{2H}\sqrt{2} \end{bmatrix}, \quad \mathbf{V}(\eta)' = \begin{bmatrix} 0 \\ 0 \end{bmatrix} \text{ for half integer } H. \tag{A5b}$$

Note that the vectors $\mathbf{V}(\alpha)'$ depend on $\mathbf{G}$. Substituting these evaluations into Eq. (128) we obtain the results for the intensities in Tables VI and VII for the out-of-plane ($\sigma=+1$) modes.

2. In-plane modes

For the in-plane sector we have (for dominant J)

$$\mathbf{A}' + \mathbf{B}' = \begin{bmatrix} x_3 + \zeta S + \frac{1}{2}\alpha & \sqrt{2}(\zeta S - \frac{1}{2}\alpha) \\ \sqrt{2}(\zeta S - \frac{1}{2}\alpha) & 2\zeta S + \alpha \end{bmatrix},$$

$$\mathbf{A}' - \mathbf{B}' = \begin{bmatrix} 8JS & \sqrt{2}J_{12}S \\ \sqrt{2}J_{12}S & 8J_2 S \end{bmatrix}, \tag{A6}$$

and we now tabulate the right eigenvectors of the block matrices $M_{-+} \equiv [\mathbf{A}' - \mathbf{B}'][\mathbf{A}' + \mathbf{B}']$ associated with the eigenvalues (the squares of the mode energies) ω_r^2. For dominant J we have the approximate results

$$\tilde{\Psi}_-^{>} = [1,0], \quad (\omega_-^{>})^2 = (8JS)(x_3 + \zeta S + \tfrac{1}{2}\alpha),$$

$$\tilde{\Psi}_-^{<} = [-\sqrt{2}(\zeta S - \tfrac{1}{2}\alpha), x_3 + \zeta S + \tfrac{1}{2}\alpha],$$

$$(\omega_-^{<})^2 = (8J_2 S)\left[2\zeta S + \alpha - 2\frac{(\zeta S - \frac{1}{2}\alpha)^2}{(x_3 + \zeta S + \frac{1}{2}\alpha)}\right] \tag{A7}$$

and

$$\mathbf{V}(z)' = \begin{bmatrix} 1+(-1)^{H+K} \\ -\sqrt{2}(-1)^{H} \end{bmatrix},$$

$$\mathbf{V}(\eta)' = \begin{bmatrix} 1-(-1)^{H+K} \\ 0 \end{bmatrix} \text{ for integer } H, \tag{A8a}$$

$$\mathbf{V}(z)' = \begin{bmatrix} 0 \\ 0 \end{bmatrix}, \quad \mathbf{V}(\eta)' = \begin{bmatrix} 0 \\ -\sqrt{2}(i)^{2H} \end{bmatrix} \text{ for half integer } H. \tag{A8b}$$

As before, only the vectors $\mathbf{V}(\alpha)'$ depend on $\mathbf{G}$. Substituting these evaluations into Eq. (129) we obtain the results for the intensities in Tables VI and VII for the in-plane ($\sigma=-1$) modes.

APPENDIX B: SHENDER PARAMETERS

In this Appendix we evaluate the averages

$$A_1 = \langle a_m f_n^\dagger \rangle, \tag{B1a}$$

$$A_2 = \langle a_m e_n \rangle, \tag{B1b}$$

where site n is a nearest neighbor of site m. The above quantities can be calculated perturbatively in the frustrated coupling J_{12} between $\mathrm{Cu_I}$'s and $\mathrm{Cu_{II}}$'s. (See Fig. 1.)

1. A_1

Thus

$$A_1 = -\left\langle 0 \left| V_{\text{I-II}} \frac{1}{\mathcal{E}} a_m f_n^\dagger \right| 0 \right\rangle - \left\langle 0 \left| a_m f_n^\dagger \frac{1}{\mathcal{E}} V_{\text{I-II}} \right| 0 \right\rangle, \tag{B2}$$

where $\mathcal{E}$ is the unperturbed energy of the virtual state relative to the ground state. Here we invoke perturbation theory relative to decoupled $\mathrm{Cu_I}$ and $\mathrm{Cu_{II}}$ subsystems, and

$$V_{\text{I-II}} = J_{12}S\Bigg[\sum_{i\in a,\delta}[a_i^\dagger a_i + e_j^\dagger e_j + a_i e_j + a_i^\dagger e_j^\dagger] + \sum_{i\in a,\delta}[a_i^\dagger a_i + f_j^\dagger f_j + a_i^\dagger f_j + f_j^\dagger a_i] + \sum_{i\in b,\delta}[b_i^\dagger b_i + e_j^\dagger e_j + b_i^\dagger e_j + e_j^\dagger b_i]$$
$$+ \sum_{i\in b,\delta}[b_i^\dagger b_i + f_j^\dagger f_j + b_i f_j + b_i^\dagger f_j^\dagger] + \sum_{i\in c,\delta}[c_i^\dagger c_i + e_j^\dagger e_j + c_i^\dagger e_j + e_j^\dagger c_i] + \sum_{i\in c,\delta}[c_i^\dagger c_i + f_j^\dagger f_j + c_i f_j + c_i^\dagger f_j^\dagger]$$
$$+ \sum_{i\in d,\delta}[d_i^\dagger d_i + e_j^\dagger e_j + d_i e_j + d_i^\dagger e_j^\dagger] + \sum_{i\in d,\delta}[d_i^\dagger d_i + f_j^\dagger f_j + d_i^\dagger f_j + f_j^\dagger d_i]\Bigg]. \tag{B3}$$

Only terms in $V_{\text{I-II}}$ which have operators in both subsystems contribute. Also, it suffices to treat each subsystem as an isotropic Heisenberg model. Accordingly, in Eq. (B2) we need keep only terms with f or $e^\dagger$ and $a^\dagger$, $d^\dagger$, b, or c. So we set

$$V_{\text{I-II}} = V_1 \equiv J_{12}S\sum_{i,\delta}[a_i^\dagger e_j^\dagger + a_i^\dagger f_j + e_j^\dagger b_i + b_i f_j + e_j^\dagger c_i + c_i f_j + d_i^\dagger e_j^\dagger + d_i^\dagger f_j]. \tag{B4}$$

Thus with $n = m + \delta_{af}$ we have $A_1 = A_1^+ + A_1^-$, where

$$A_1^+ = -J_{12}S\sum_{i\in a}\left\langle 0\left|a_i^\dagger\frac{1}{\mathcal{E}}a_m\right|0\right\rangle\langle 0|[e_{i+\delta_{ae}}^\dagger + f_{i+\delta_{af}}]f_{m+\delta_{af}}^\dagger|0\rangle - J_{12}S\sum_{i\in b}\left\langle 0\left|b_i\frac{1}{\mathcal{E}}a_m\right|0\right\rangle\langle 0|[e_{i+\delta_{be}}^\dagger + f_{i+\delta_{bf}}]f_{m+\delta_{af}}^\dagger|0\rangle$$
$$- J_{12}S\sum_{i\in c}\left\langle 0\left|c_i\frac{1}{\mathcal{E}}a_m\right|0\right\rangle\langle 0|[e_{i+\delta_{ce}}^\dagger + f_{i+\delta_{cf}}]f_{m+\delta_{af}}^\dagger|0\rangle - J_{12}S\sum_{i\in d}\left\langle 0\left|d_i^\dagger\frac{1}{\mathcal{E}}a_m\right|0\right\rangle\langle 0|[e_{i+\delta_{de}}^\dagger + f_{i+\delta_{df}}]f_{m+\delta_{af}}^\dagger|0\rangle, \tag{B5a}$$

$$A_1^- = -J_{12}S\sum_{i\in a}\left\langle 0\left|a_m\frac{1}{\mathcal{E}}a_i^\dagger\right|0\right\rangle\langle 0|f_{m+\delta_{af}}^\dagger[e_{i+\delta_{ae}}^\dagger + f_{i+\delta_{af}}]|0\rangle - J_{12}S\sum_{i\in b}\left\langle 0\left|a_m\frac{1}{\mathcal{E}}b_i\right|0\right\rangle\langle 0|f_{m+\delta_{af}}^\dagger[e_{i+\delta_{be}}^\dagger + f_{i+\delta_{bf}}]|0\rangle$$
$$- J_{12}S\sum_{i\in c}\left\langle 0\left|a_m\frac{1}{\mathcal{E}}c_i\right|0\right\rangle\langle 0|f_{m+\delta_{af}}^\dagger[e_{i+\delta_{ce}}^\dagger + f_{i+\delta_{cf}}]|0\rangle - J_{12}S\sum_{i\in d}\left\langle 0\left|a_m\frac{1}{\mathcal{E}}d_i^\dagger\right|0\right\rangle\langle 0|f_{m+\delta_{af}}^\dagger[e_{i+\delta_{de}}^\dagger + f_{i+\delta_{df}}]|0\rangle. \tag{B5b}$$

Here we neglected the energy of the Cu_{II} modes in comparison to that of the Cu_I modes. Also we used the unusual notation that

$$\boldsymbol{\delta}_{st} = \mathbf{r}_t - \mathbf{r}_s\,. \tag{B6}$$

As will become clearer as we proceed, one can deduce the form of A_1^- from that of A_1^+ by interchanging the l and m coefficients defined in Eqs. (B9) and (B16), below. Therefore we focus on A_1^+ which is

$$A_1^+ = -\frac{J_{12}S}{N_{\mathrm{uc}}^2}\sum_{\mathbf{q},\mathbf{k}}\sum_{i\in a}\left\langle 0\left|a^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle e^{i(\mathbf{q}+\mathbf{k})\cdot\mathbf{r}_{im}}\langle 0|[e^\dagger(\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{ae}} + f(-\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{af}}]f^\dagger(-\mathbf{k})e^{-i\mathbf{k}\cdot\boldsymbol{\delta}_{af}}|0\rangle$$
$$-\frac{J_{12}S}{N_{\mathrm{uc}}^2}\sum_{\mathbf{q},\mathbf{k}}\sum_{i\in b}\left\langle 0\left|b(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle e^{i(\mathbf{q}+\mathbf{k})\cdot\mathbf{r}_{im}}\langle 0|[e^\dagger(\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{be}} + f(-\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{bf}}]f^\dagger(-\mathbf{k})e^{-i\mathbf{k}\cdot\boldsymbol{\delta}_{af}}|0\rangle$$
$$-\frac{J_{12}S}{N_{\mathrm{uc}}^2}\sum_{\mathbf{q},\mathbf{k}}\sum_{i\in c}\left\langle 0\left|c(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle e^{i(\mathbf{q}+\mathbf{k})\cdot\mathbf{r}_{im}}\langle 0|[e^\dagger(\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{ce}} + f(-\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{cf}}]f^\dagger(-\mathbf{k})e^{-i\mathbf{k}\cdot\boldsymbol{\delta}_{af}}]|0\rangle$$
$$-\frac{J_{12}S}{N_{\mathrm{uc}}^2}\sum_{\mathbf{q},\mathbf{k}}\sum_{i\in d}\left\langle 0\left|d^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle e^{i(\mathbf{q}+\mathbf{k})\cdot\mathbf{r}_{im}}\langle 0|[e^\dagger(\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{de}} + f(-\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{df}}]f^\dagger(-\mathbf{k})e^{-i\mathbf{k}\cdot\boldsymbol{\delta}_{af}}|0\rangle. \tag{B7}$$

Doing the sum over i we get

$$A_1^+ = -\frac{J_{12}S}{N_{\mathrm{uc}}}\sum_{\mathbf{q}}\left\langle 0\left|a^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle\langle 0|[e^\dagger(-\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{ae}} + f(\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{af}}]f^\dagger(\mathbf{q})e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{af}}|0\rangle$$
$$-\frac{J_{12}S}{N_{\mathrm{uc}}}\sum_{\mathbf{q}}\left\langle 0\left|b(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle\langle 0|[e^\dagger(-\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{be}} + f(\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{bf}}]f^\dagger(\mathbf{q})e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{af}}|0\rangle$$
$$-\frac{J_{12}S}{N_{\mathrm{uc}}}\sum_{\mathbf{q}}\left\langle 0\left|c(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle\langle 0|[e^\dagger(-\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{ce}} + f(\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{cf}}]f^\dagger(\mathbf{q})e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{af}}]|0\rangle$$
$$-\frac{J_{12}S}{N_{\mathrm{uc}}}\sum_{\mathbf{q}}\left\langle 0\left|d^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle\langle 0|[e^\dagger(-\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{de}} + f(\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{df}}]f^\dagger(\mathbf{q})e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{af}}|0\rangle. \tag{B8}$$

For the Cu_{II} subsystem we have the usual relations

$$e(\mathbf{q}) = l_{\mathbf{q}}\eta_{\mathbf{q}} - m_{\mathbf{q}}\delta_{-\mathbf{q}}^\dagger, \quad f^\dagger(-\mathbf{q}) = -m_{\mathbf{q}}\eta_{\mathbf{q}} + l_{\mathbf{q}}\delta_{-\mathbf{q}}^\dagger, \tag{B9}$$

where $\eta(\mathbf{q})$ and $\delta(\mathbf{q})$ are the normal mode operators for the Cu_{II} subsystem and $l_{\mathbf{q}}$ and $m_{\mathbf{q}}$ are given by

$$l_{\mathbf{q}}^2 = \frac{1+\epsilon(\mathbf{q})}{2\epsilon(\mathbf{q})}, \quad m_{\mathbf{q}}^2 = \frac{1-\epsilon(\mathbf{q})}{2\epsilon(\mathbf{q})}, \quad l_{\mathbf{q}}m_{\mathbf{q}} = -\frac{\gamma(\mathbf{q})}{2\epsilon(\mathbf{q})}, \tag{B10}$$

where $\gamma(\mathbf{q}) = \frac{1}{2}[\cos(aq_x) + \cos(aq_y)]$ and $\epsilon(\mathbf{q})^2 = 1 - \gamma(\mathbf{q})^2$. In the ground state we evaluate the averages to get

$$A_1^+ = -\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|a^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle[-l_{\mathbf{q}}m_{\mathbf{q}}e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{ae}}+l_{\mathbf{q}}^2e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{af}}]e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{af}} - \frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|b(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle$$
$$\times[-l_{\mathbf{q}}m_{\mathbf{q}}e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{be}}+l_{\mathbf{q}}^2e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{bf}}]e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{af}} - \frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|c(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle$$
$$\times[-l_{\mathbf{q}}m_{\mathbf{q}}e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{ce}}+l_{\mathbf{q}}^2e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{cf}}]e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{af}} - \frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|d^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle[-l_{\mathbf{q}}m_{\mathbf{q}}e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{de}}+l_{\mathbf{q}}^2e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{df}}]e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{af}}$$
$$= -\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|a^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle[-l_{\mathbf{q}}m_{\mathbf{q}}e^{iaq_x}+l_{\mathbf{q}}^2] - \frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|b(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle[-l_{\mathbf{q}}m_{\mathbf{q}}e^{-iq_ya/2}$$
$$+l_{\mathbf{q}}^2e^{iq_ya/2}]e^{iq_xa/2} - \frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|c(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle[-l_{\mathbf{q}}m_{\mathbf{q}}e^{iq_ya/2}+l_{\mathbf{q}}^2e^{-iq_ya/2}]e^{iq_xa/2}$$
$$-\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|d^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle[-l_{\mathbf{q}}m_{\mathbf{q}}+l_{\mathbf{q}}^2e^{iq_xa}]. \tag{B11}$$

For the $\mathrm{Cu_I}$ subsystem we have normal modes via the transformations

$$a(\mathbf{q}) = (1/\sqrt{2})[a_+(\mathbf{q})+a_-(\mathbf{q})],\quad d(\mathbf{q}) = (1/\sqrt{2})[a_+(\mathbf{q})-a_-(\mathbf{q})],$$
$$b(\mathbf{q}) = (1/\sqrt{2})[b_+(\mathbf{q})+b_-(\mathbf{q})],\quad c(\mathbf{q}) = (1/\sqrt{2})[b_+(\mathbf{q})-b_-(\mathbf{q})]. \tag{B12}$$

In terms of these operators (in the order a_+, b_+, a_-, b_-) we have the matrices **A** and **B**:

$$\frac{\mathbf{A}(\mathbf{q})}{S} = \begin{bmatrix} 4J+2J_3 & 0 & 0 & 0 \\ 0 & 4J+2J_3 & 0 & 0 \\ 0 & 0 & 4J+2J_3 & 0 \\ 0 & 0 & 0 & 4J+2J_3 \end{bmatrix} \tag{B13}$$

and $\mathbf{B}(\mathbf{q})/S$ as

$$\begin{bmatrix} 0 & 2J(c_++c_-)+2J_3c_z & 0 & 0 \\ 2J(c_++c_-)+2J_3c_z & 0 & 0 & 0 \\ 0 & 0 & 0 & 2J(c_+-c_-)+2J_3c_z \\ 0 & 0 & 2J(c_+-c_-)+2J_3c_z & 0 \end{bmatrix}, \tag{B14}$$

where

$$c_+ = \cos[a(q_x+q_y)/2],\quad c_- = \cos[a(q_x-q_y)/2],$$
$$c_z = \cos(q_zc/2). \tag{B15}$$

Now each sector has relations analogous to the $\mathrm{Cu_I}$'s:

$$a_\sigma(\mathbf{q}) = l_{\sigma,\mathbf{q}}\alpha_\sigma(\mathbf{q}) - m_{\sigma,\mathbf{q}}\beta_\sigma^\dagger(-\mathbf{q}),$$
$$b_\sigma^\dagger(-\mathbf{q}) = -m_{\sigma,\mathbf{q}}\alpha_\sigma(\mathbf{q}) + l_{\sigma,\mathbf{q}}\beta_\sigma^\dagger(-\mathbf{q}), \tag{B16}$$

where $\alpha_\sigma(\mathbf{q})$ and $\beta_\sigma(\mathbf{q})$ are the normal mode operators (with $\sigma=+$ or $\sigma=-$), and

$$l_{\sigma,\mathbf{q}}^2 = \frac{A+E_\sigma(\mathbf{q})}{2E_\sigma(\mathbf{q})},\quad m_{\sigma,\mathbf{q}}^2 = \frac{A-E_\sigma(\mathbf{q})}{2E_\sigma(\mathbf{q})},$$

$$l_{\sigma,\mathbf{q}}m_{\sigma,\mathbf{q}} = \frac{B_\sigma(\mathbf{q})}{2E_\sigma(\mathbf{q})}. \tag{B17}$$

Here

$$E_\sigma(\mathbf{q})^2 = A^2 - B_\sigma(\mathbf{q})^2, \tag{B18}$$

where

$$A = 4JS + 2J_3S,$$

$$B_\sigma(\mathbf{q}) = 2JS(\cos[(q_x+q_y)a/2] + \sigma\cos[(q_x-q_y)a/2])$$
$$+2J_3S\cos(q_zc). \tag{B19}$$

Thus

$$\left\langle 0\left|a^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle = \tfrac{1}{2}\sum_\sigma \langle 0|a_\sigma^\dagger(\mathbf{q})a_\sigma(\mathbf{q})|0\rangle E_\sigma(\mathbf{q})^{-1} = \tfrac{1}{2}\sum_\sigma m_{\sigma\mathbf{q}}^2 E_\sigma(\mathbf{q})^{-1}$$
$$= \sum_\sigma \frac{A-E_\sigma(\mathbf{q})}{4E_\sigma(\mathbf{q})^2}. \tag{B20}$$

Similarly

$$\left\langle 0\left|a(\mathbf{q})\frac{1}{\mathcal{E}}a^\dagger(\mathbf{q})\right|0\right\rangle = \tfrac{1}{2}\sum_\sigma l_{\sigma\mathbf{q}}^2 E_\sigma(\mathbf{q})^{-1} = \sum_\sigma \frac{A+E_\sigma(\mathbf{q})}{4E_\sigma(\mathbf{q})^2}, \tag{B21}$$

$$\left\langle 0\left|d^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle = \tfrac{1}{2}\sum_\sigma \sigma m_{\sigma\mathbf{q}}^2 E_\sigma(\mathbf{q})^{-1} = \sum_\sigma \sigma\frac{A-E_\sigma(\mathbf{q})}{4E_\sigma(\mathbf{q})^2}, \tag{B22}$$

$$\left\langle 0\left|a(\mathbf{q})\frac{1}{\mathcal{E}}d^\dagger(\mathbf{q})\right|0\right\rangle = \tfrac{1}{2}\sum_\sigma \sigma l_{\sigma\mathbf{q}}^2 E_\sigma(\mathbf{q})^{-1} = \sum_\sigma \sigma\frac{A+E_\sigma(\mathbf{q})}{4E_\sigma(\mathbf{q})^2}, \tag{B23}$$

$$\left\langle 0\left|b(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle = -\tfrac{1}{2}\sum_\sigma l_{\sigma,\mathbf{q}}m_{\sigma\mathbf{q}} E_\sigma(\mathbf{q})^{-1} = -\sum_\sigma \frac{B_\sigma(\mathbf{q})}{4E_\sigma(\mathbf{q})^2}, \tag{B24}$$

$$\left\langle a\left|(\mathbf{q})\frac{1}{\mathcal{E}}b(-\mathbf{q})\right|0\right\rangle = -\tfrac{1}{2}\sum_\sigma l_{\sigma,\mathbf{q}}m_{\sigma\mathbf{q}} E_\sigma(\mathbf{q})^{-1} = -\sum_\sigma \frac{B_\sigma(\mathbf{q})}{4E_\sigma(\mathbf{q})^2}, \tag{B25}$$

$$\left\langle 0\left|c(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle = -\tfrac{1}{2}\sum_\sigma \sigma l_{\sigma,\mathbf{q}}m_{\sigma\mathbf{q}} E_\sigma(\mathbf{q})^{-1} = -\sum_\sigma \sigma\frac{B_\sigma(\mathbf{q})}{4E_\sigma(\mathbf{q})^2}, \tag{B26}$$

$$\left\langle 0\left|a(\mathbf{q})\frac{1}{\mathcal{E}}c(-\mathbf{q})\right|0\right\rangle = -\tfrac{1}{2}\sum_\sigma \sigma l_{\sigma,\mathbf{q}}m_{\sigma\mathbf{q}} E_\sigma(\mathbf{q})^{-1} = -\sum_\sigma \sigma\frac{B_\sigma(\mathbf{q})}{4E_\sigma(\mathbf{q})^2}. \tag{B27}$$

Then

$$\begin{aligned} A_1 &= A_1^+ + A_1^- \\ &= -\frac{J_{12}S}{8N_{\rm uc}}\sum_{\mathbf{q}}\sum_\sigma [E_\sigma(\mathbf{q})^2\epsilon(\mathbf{q})]^{-1}\{[A-E_\sigma(\mathbf{q})][-\gamma(\mathbf{q})e^{iq_xa}+1+\epsilon(\mathbf{q})]+[A+E_\sigma(\mathbf{q})][-\gamma(\mathbf{q})e^{iq_xa}+1-\epsilon(\mathbf{q})] \\ &\quad -B_\sigma(\mathbf{q})[-\gamma(\mathbf{q})e^{-iq_ya/2}+(1+\epsilon(\mathbf{q}))e^{iq_ya/2}]e^{iq_xa/2}-B_\sigma(\mathbf{q})[-\gamma(\mathbf{q})e^{-iq_ya/2}+(1-\epsilon(\mathbf{q}))e^{iq_ya/2}]e^{iq_xa/2}-\sigma B_\sigma(\mathbf{q}) \\ &\quad \times[-\gamma(\mathbf{q})e^{iq_ya/2}+(1+\epsilon(\mathbf{q}))e^{-iq_ya/2}]e^{iq_xa/2}-\sigma B_\sigma(\mathbf{q})[-\gamma(\mathbf{q})e^{iq_ya/2}+(1-\epsilon(\mathbf{q}))e^{-iq_ya/2}]e^{iq_xa/2} \\ &\quad +\sigma[A-E_\sigma(\mathbf{q})][-\gamma(\mathbf{q})+(1+\epsilon(\mathbf{q}))e^{iq_xa}]+\sigma[A+E_\sigma(\mathbf{q})][-\gamma(\mathbf{q})+(1-\epsilon(\mathbf{q}))e^{iq_xa}]\}, \end{aligned} \tag{B28}$$

where

$$\gamma(\mathbf{q}) = \tfrac{1}{2}[\cos(q_xa)+\cos(q_ya)] \tag{B29}$$

and

$$\epsilon(\mathbf{q})^2 = 1-\gamma(\mathbf{q})^2. \tag{B30}$$

We use the fact that $J_3 \ll J$. Only if a sum is divergent will it make a difference if we retain nonzero J_3. So we tentatively assume no divergences and write

$$B_+(\mathbf{q}) = 4JS\cos(q_xa/2)\cos(q_ya/2), \tag{B31a}$$

$$B_-(\mathbf{q}) = -4JS\sin(q_xa/2)\sin(q_ya/2). \tag{B31b}$$

We now simplify Eq. (B28). We note that under the sum over wavevectors we can replace $\exp(iq_xa)$ by $\gamma(\mathbf{q})$. Let us apply the same reasoning to $\exp[i(q_x\pm q_y)a/2]$:

$$\begin{aligned} &\exp[i(q_x\pm q_y)a/2] \\ &= \cos(q_xa/2)\cos(q_ya/2)\mp\sin(q_xa/2)\sin(q_ya/2) \\ &\quad + i[\sin(q_xa/2)\cos(q_ya/2)a\pm\cos(q_xa/2)\sin(q_ya/2)]. \end{aligned} \tag{B32}$$

After summation over wave vectors the imaginary parts will drop out. So

$$\exp[i(q_x+q_y)a/2]$$
$$\Rightarrow \cos(q_xa/2)\cos(q_ya/2)-\sin(q_xa/2)\sin(q_ya/2)$$
$$=\left(\frac{1}{4JS}\right)\sum_\sigma B_\sigma(\mathbf{q}), \tag{B33a}$$

$$\exp[i(q_x-q_y)a/2]$$
$$\Rightarrow \cos(q_xa/2)\cos(q_ya/2)+\sin(q_xa/2)\sin(q_ya/2)$$
$$=\left(\frac{1}{4JS}\right)\sum_\sigma \sigma B_\sigma(\mathbf{q}). \tag{B33b}$$

In this connection note that sums which are proportional to $B_+(\mathbf{q})B_-(\mathbf{q})$ vanish. So

$$A_1=-\frac{J_{12}S}{8N_{\rm uc}}\sum_{\mathbf{q}}\sum_\sigma\left(\frac{1}{E_\sigma(\mathbf{q})^2\epsilon(\mathbf{q})}\right)\Big\{[A-E_\sigma(\mathbf{q})][-\gamma(\mathbf{q})^2+1+\epsilon(\mathbf{q})]+[A+E_\sigma(\mathbf{q})][-\gamma(\mathbf{q})^2+1-\epsilon(\mathbf{q})]+\sigma[A-E_\sigma(\mathbf{q})]$$
$$\times[\gamma(\mathbf{q})\epsilon(\mathbf{q})]+\sigma[A+E_\sigma(\mathbf{q})][-\gamma(\mathbf{q})\epsilon(\mathbf{q})]-\left(\frac{B_\sigma(\mathbf{q})}{4JS}\right)[-\gamma(\mathbf{q})\sigma B_\sigma(\mathbf{q})+(1+\epsilon(\mathbf{q}))B_\sigma(\mathbf{q})]$$
$$-\left(\frac{B_\sigma(\mathbf{q})}{4JS}\right)[-\gamma(\mathbf{q})\sigma B_\sigma(\mathbf{q})+(1-\epsilon(\mathbf{q}))B_\sigma(\mathbf{q})]-\left(\frac{\sigma B_\sigma(\mathbf{q})}{4JS}\right)[-\gamma(\mathbf{q})B_\sigma(\mathbf{q})+(1+\epsilon(\mathbf{q}))\sigma B_\sigma(\mathbf{q})]$$
$$-\left(\frac{\sigma B_\sigma(\mathbf{q})}{4JS}\right)[-\gamma(\mathbf{q})B_\sigma(\mathbf{q})+(1-\epsilon(\mathbf{q}))\sigma B_\sigma(\mathbf{q})]\Big\}$$
$$=-\frac{J_{12}S}{8N_{\rm uc}}\sum_{\mathbf{q}}\sum_\sigma\left(\frac{1}{E_\sigma(\mathbf{q})^2\epsilon(\mathbf{q})}\right)\{2A\epsilon(\mathbf{q})^2-2E_\sigma(\mathbf{q})\epsilon(\mathbf{q})-2E_\sigma(\mathbf{q})\epsilon(\mathbf{q})\sigma\gamma(\mathbf{q})-(JS)^{-1}B_\sigma(\mathbf{q})^2[1-\sigma\gamma(\mathbf{q})]\}. \tag{B34}$$

Now we must understand how the wave vector sums are to be done. The unit cell is

$$\mathbf{a}_1=a\hat{x}+a\hat{y},\quad \mathbf{a}_2=-a\hat{x}+a\hat{y}. \tag{B35}$$

Thus the reciprocal lattice vectors are

$$\mathbf{G}_1=(\pi/a)(\hat{x}+\hat{y}),\quad \mathbf{G}_2=(\pi/a)(-\hat{x}+\hat{y}). \tag{B36}$$

Thus the sums are carried over the first zone, shown below in Fig. 7.

2. A_2

Thus

$$A_2=-\left\langle 0\left|V_{\text{I-II}}\frac{1}{\mathcal{E}}a_me_n\right|0\right\rangle-\left\langle 0\left|a_me_n\frac{1}{\mathcal{E}}V_{\text{I-II}}\right|0\right\rangle, \tag{B37}$$

where we invoke perturbation theory relative to decoupled and isotropic $\mathrm{Cu_I}$ and $\mathrm{Cu_{II}}$ subsystems. As for A_1 effectively we have Eq. (B4). Thus, as before, we write $A_2=A_2^++A_2^-$, where A_2^- is obtained from A_2^+ by interchanging all l and m coefficients and

$$A_2^+=-J_{12}S\sum_{i\in a}\left\langle 0\left|a_i^\dagger\frac{1}{\mathcal{E}}a_m\right|0\right\rangle\langle 0|[e^\dagger_{i+\delta_{ae}}+f_{i+\delta_{af}}]e_{m+\delta_{ae}}|0\rangle-J_{12}S\sum_{i\in b}\left\langle 0\left|b_i\frac{1}{\mathcal{E}}a_m\right|0\right\rangle\langle 0|[e^\dagger_{i+\delta_{be}}+f_{i+\delta_{bf}}]e_{m+\delta_{ae}}|0\rangle$$
$$-J_{12}S\sum_{i\in c}\left\langle 0\left|c_i\frac{1}{\mathcal{E}}a_m\right|0\right\rangle\langle 0|[e^\dagger_{i+\delta_{ce}}+f_{i+\delta_{cf}}]e_{m+\delta_{ae}}|0\rangle-J_{12}S\sum_{i\in d}\left\langle 0\left|d_i^\dagger\frac{1}{\mathcal{E}}a_m\right|0\right\rangle\langle 0|[e^\dagger_{i+\delta_{de}}+f_{i+\delta_{df}}]e_{m+\delta_{ae}}|0\rangle. \tag{B38}$$

Then

$$A_2^+ = -\frac{J_{12}S}{N_{\rm uc}^2}\sum_{\mathbf{q},\mathbf{k}}\sum_{i\in a}\left\langle 0\left|a^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle e^{i(\mathbf{q}+\mathbf{k})\cdot\mathbf{r}_{im}}\langle 0|[e^\dagger(\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{ae}}+f(-\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{af}}]e(\mathbf{k})e^{-i\mathbf{k}\cdot\boldsymbol{\delta}_{ae}}|0\rangle$$
$$-\frac{J_{12}S}{N_{\rm uc}^2}\sum_{\mathbf{q},\mathbf{k}}\sum_{i\in b}\left\langle 0\left|b(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle e^{i(\mathbf{q}+\mathbf{k})\cdot\mathbf{r}_{im}}\langle 0|[e^\dagger(\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{be}}+f(-\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{bf}}]e(\mathbf{k})e^{-i\mathbf{k}\cdot\boldsymbol{\delta}_{ae}}|0\rangle$$
$$-\frac{J_{12}S}{N_{\rm uc}^2}\sum_{\mathbf{q},\mathbf{k}}\sum_{i\in c}\left\langle 0\left|c(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle e^{i(\mathbf{q}+\mathbf{k})\cdot\mathbf{r}_{im}}\langle 0|[e^\dagger(\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{ce}}+f(-\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{cf}}]e(\mathbf{k})e^{-i\mathbf{k}\cdot\boldsymbol{\delta}_{ae}}]|0\rangle$$
$$-\frac{J_{12}S}{N_{\rm uc}^2}\sum_{\mathbf{q},\mathbf{k}}\sum_{i\in d}\left\langle 0\left|d^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle e^{i(\mathbf{q}+\mathbf{k})\cdot\mathbf{r}_{im}}\langle 0|[e^\dagger(\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{de}}+f(-\mathbf{k})e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{df}}]e(\mathbf{k})e^{-i\mathbf{k}\cdot\boldsymbol{\delta}_{ae}}|0\rangle. \quad \text{(B39)}$$

Doing the sum over i we get

$$A_2^+ = -\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|a^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle\langle 0|[e^\dagger(-\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{ae}}+f(\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{af}}]e(-\mathbf{q})e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{ae}}|0\rangle$$
$$-\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|b(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle\langle 0|[e^\dagger(-\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{be}}+f(\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{bf}}]e(-\mathbf{q})e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{ae}}|0\rangle$$
$$-\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|c(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle\langle 0|[e^\dagger(-\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{ce}}+f(\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{cf}}]e(-\mathbf{q})e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{ae}}]|0\rangle$$
$$-\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|d^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle\langle 0|[e^\dagger(-\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{de}}+f(\mathbf{q})e^{-i\mathbf{q}\cdot\boldsymbol{\delta}_{df}}]e(-\mathbf{q})e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{ae}}|0\rangle. \quad \text{(B40)}$$

This is

$$A_2^+ = -\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|a^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle[\langle 0|e^\dagger(-\mathbf{q})e(-\mathbf{q})|0\rangle+\langle 0|f(\mathbf{q})e(-\mathbf{q})|0\rangle e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{fae}}]$$
$$-\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|b(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle\Big[\langle 0|[e^\dagger(-\mathbf{q})e(-\mathbf{q})|0\rangle e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{aeb}}+\langle 0|f(\mathbf{q})e(-\mathbf{q})|0\rangle e^{i\mathbf{q}\cdot(\boldsymbol{\delta}_{ae}-\boldsymbol{\delta}_{bf})}]$$
$$-\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|c(-\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle[\langle 0|[e^\dagger(-\mathbf{q})e(-\mathbf{q})|0\rangle e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{aec}}+\langle 0|f(\mathbf{q})e(-\mathbf{q})|0\rangle e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{ae}-i\mathbf{q}\cdot\boldsymbol{\delta}_{cf}}]$$
$$-\frac{J_{12}S}{N_{\rm uc}}\sum_{\mathbf{q}}\left\langle 0\left|d^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}a(\mathbf{q})\right|0\right\rangle[\langle 0|[e^\dagger(-\mathbf{q})e(-\mathbf{q})|0\rangle e^{i\mathbf{q}\cdot\boldsymbol{\delta}_{aed}}+\langle 0|f(\mathbf{q})e(-\mathbf{q})|0\rangle e^{i\mathbf{q}\cdot(\boldsymbol{\delta}_{ae}-\boldsymbol{\delta}_{df})}]. \quad \text{(B41)}$$

Here the symbol δ_{fae} denotes the vector which goes from an f site to an e site via an a site, such that fae is a sequence of nearest-neighboring sites. So

$$A_2 = A_2^+ + A_2^-$$
$$= -\frac{J_{12}S}{4N_{\rm uc}}\sum_{\mathbf{q}}\sum_{\sigma}\{E_\sigma(\mathbf{q})^{-2}[A-E_\sigma(\mathbf{q})][m_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}e^{-iq_xa}]+[A+E_\sigma(\mathbf{q})][l_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}e^{-iq_xa}]$$
$$-B_\sigma(\mathbf{q})[e^{-i(q_x+q_y)a/2}m_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}e^{i(q_y-q_x)a/2}]-B_\sigma(\mathbf{q})[e^{-i(q_x+q_y)a/2}l_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}e^{i(q_y-q_x)a/2}]$$
$$-\sigma B_\sigma(\mathbf{q})[e^{i(q_y-q_x)a/2}m_{\mathbf{q}}^2-e^{-i(q_x+q_y)a/2}l_{\mathbf{q}}m_{\mathbf{q}}]-\sigma B_\sigma(\mathbf{q})[e^{i(q_y-q_x)a/2}l_{\mathbf{q}}^2-e^{-i(q_x+q_y)a/2}l_{\mathbf{q}}m_{\mathbf{q}}]$$
$$+\sigma[A-E_\sigma][e^{-iq_xa}m_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}]+\sigma[A+E_\sigma][e^{-iq_xa}m_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}]\}. \quad \text{(B42)}$$

Making the same replacements as in A_1 we get

$$A_2=-\frac{J_{12}S}{4N_{\rm uc}}\sum_{\mathbf{q}}\sum_{\sigma} E_\sigma(\mathbf{q})^{-2}\{[A-E_\sigma(\mathbf{q})][m_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}\gamma(\mathbf{q})]+[A+E_\sigma(\mathbf{q})][l_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}\gamma(\mathbf{q})]$$
$$-B_\sigma(\mathbf{q})B_\sigma(\mathbf{q})(4JS)^{-1}[m_{\mathbf{q}}^2-\sigma l_{\mathbf{q}}m_{\mathbf{q}}]-B_\sigma(\mathbf{q})B_\sigma(\mathbf{q})(4JS)^{-1}[l_{\mathbf{q}}^2-\sigma l_{\mathbf{q}}m_{\mathbf{q}}]-\sigma B_\sigma(\mathbf{q})B_\sigma(\mathbf{q})(4JS)^{-1}[\sigma m_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}]$$
$$-\sigma B_\sigma(\mathbf{q})B_\sigma(\mathbf{q})(4JS)^{-1}[\sigma l_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}]+\sigma[A-E_\sigma][\gamma(\mathbf{q})m_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}]+\sigma[A+E_\sigma][\gamma(\mathbf{q})l_{\mathbf{q}}^2-l_{\mathbf{q}}m_{\mathbf{q}}]\}. \tag{B43}$$

This is

$$A_2=-\frac{J_{12}S}{8N_{\rm uc}}\sum_{\mathbf{q}}\sum_{\sigma} E_\sigma(\mathbf{q})^{-2}\epsilon(\mathbf{q})^{-1}\Bigg[[A-E_\sigma(\mathbf{q})][1-\epsilon(\mathbf{q})-\gamma(\mathbf{q})^2]+[A+E_\sigma(\mathbf{q})][1+\epsilon(\mathbf{q})-\gamma(\mathbf{q})^2]$$
$$-\left(\frac{B_\sigma(\mathbf{q})^2}{4JS}\right)[2-2\sigma\gamma(\mathbf{q})]-\left(\frac{\sigma B_\sigma(\mathbf{q})^2}{4JS}\right)[2\sigma-2\gamma(\mathbf{q})]+\sigma[A-E_\sigma(\mathbf{q})]\{\gamma(\mathbf{q})[1-\epsilon(\mathbf{q})]-\gamma(\mathbf{q})\}$$
$$+\sigma[A+E_\sigma(\mathbf{q})]\{\gamma(\mathbf{q})[1+\epsilon(\mathbf{q})]+\gamma(\mathbf{q})\}\Bigg]. \tag{B44}$$

So

$$A_2=-\frac{J_{12}S}{8N_{\rm uc}}\sum_{\mathbf{q}}\sum_{\sigma}\left(\frac{1}{E_\sigma(\mathbf{q})^2\epsilon(\mathbf{q})}\right)\{2A\epsilon(\mathbf{q})^2+2E_\sigma(\mathbf{q})\epsilon(\mathbf{q})+2E_\sigma(\mathbf{q})\sigma\gamma(\mathbf{q})\epsilon(\mathbf{q})-B_\sigma(\mathbf{q})^2(JS)^{-1}[1-\sigma\gamma(\mathbf{q})]\}. \tag{B45}$$

3. Summary

So

$$A_1=-\left(\frac{J_{12}}{2J}\right)(C_\alpha-C_\beta) \tag{B46a}$$

$$A_2=-\left(\frac{J_{12}}{2J}\right)(C_\alpha+C_\beta), \tag{B46b}$$

where

$$C_\alpha=\frac{JS}{4N_{\rm uc}}\sum_{\mathbf{q}}\sum_{\sigma}\left(\frac{1}{E_\sigma(\mathbf{q})^2\epsilon(\mathbf{q})}\right)\{2A\epsilon(\mathbf{q})^2-B_\sigma(\mathbf{q})^2(JS)^{-1}[1-\sigma\gamma(\mathbf{q})]\} \tag{B47a}$$

$$C_\beta=\frac{JS}{4N_{\rm uc}}\sum_{\mathbf{q}}\sum_{\sigma}\left(\frac{1}{E_\sigma(\mathbf{q})}\right)\{2[1+\sigma\gamma(\mathbf{q})]\}. \tag{B47b}$$

If we extend the sum over $-\pi/a<q_x,q_y<\pi/a$, then we may write these as

$$C_\alpha=\frac{JS}{4N_{\rm uc}}\sum_{\mathbf{q}}\left(\frac{1}{E_+(\mathbf{q})^2\epsilon(\mathbf{q})}\right)\{2A\epsilon(\mathbf{q})^2-B_+(\mathbf{q})^2(JS)^{-1}[1-\gamma(\mathbf{q})]\} \tag{B48a}$$

$$C_\beta=\frac{JS}{4N_{\rm uc}}\sum_{\mathbf{q}}\left(\frac{1}{E_+(\mathbf{q})}\right)\{2[1+\gamma(\mathbf{q})]\}. \tag{B48b}$$

Of course, note that now $\Sigma_{\mathbf{q}}=2N_{uc}$. So it is convenient to introduce the notation $\langle\cdots\rangle_{\mathbf{q}}$ to denote $(2N_{uc})^{-1}\Sigma_{\mathbf{q}}$. Then

$$C_\alpha=\frac{JS}{2}\left\langle\left(\frac{1}{E_+(\mathbf{q})^2\epsilon(\mathbf{q})}\right)\{2A\epsilon(\mathbf{q})^2-B_+(\mathbf{q})^2(JS)^{-1}[1-\gamma(\mathbf{q})]\}\right\rangle_{\mathbf{q}}, \tag{B49a}$$

$$C_\beta=\frac{JS}{2}\left\langle\left(\frac{1}{E_+(\mathbf{q})}\right)\{2[1+\gamma(\mathbf{q})]\}\right\rangle_{\mathbf{q}} \tag{B49b}$$

or

$$C_\alpha=\frac{1}{4}\left\langle\frac{1-\gamma(\mathbf{q})^2-2\cos^2(aq_x/2)\cos^2(aq_y/2)[1-\gamma(\mathbf{q})]}{[1-\cos^2(aq_x/2)\cos^2(aq_y/2)]\sqrt{1-\gamma(\mathbf{q})^2}}\right\rangle_{\mathbf{q}} \tag{B50a}$$

$$C_\beta=\frac{1}{4}\left\langle\frac{1+\gamma(\mathbf{q})}{\sqrt{1-\cos^2(aq_x/2)\cos^2(aq_y/2)}}\right\rangle_{\mathbf{q}}. \tag{B50b}$$

In the approximation that $\gamma(\mathbf{q})=0$, etc. $C_\alpha=C_\beta=\frac{1}{4}$. Numerical evaluation yields

$$C_\alpha=0.1686, \quad C_\beta=0.4210. \tag{B51}$$

APPENDIX C: IN-PLANE Cu_I-Cu_I INTERACTION

Here we reproduce by perturbation theory the gap found phenomenologically by Yildirim *et al.*[13] We treat an antiferromagnet on a square lattice (of lattice constant a), in which there are two sublattices, a and b. The lattice is shown in the Fig. 6 with the magnetic unit cell within dashed lines. The magnetic unit cell has basis vectors

$$\mathbf{a}_1=a\hat{\xi}+a\hat{\eta},$$

$$\mathbf{a}_2=-a\hat{\xi}+a\hat{\eta}. \tag{C1}$$

We transform to bosons using Eq. (13).

First we consider terms $\mathcal{H}$ in the Hamiltonian which are quadratic in boson operators. We write

$$\mathcal{H}=\mathcal{H}_J+\mathcal{H}_\delta. \tag{C2}$$

Here

$$\mathcal{H}_J=4JS\sum_{\mathbf{q}}\{a^\dagger(\mathbf{q})a(\mathbf{q})+b^\dagger(\mathbf{q})b(\mathbf{q})+\gamma(\mathbf{q})\times[a^\dagger(\mathbf{q})b^\dagger(-\mathbf{q})+a(\mathbf{q})b(-\mathbf{q})]\}, \tag{C3}$$

with

$$\gamma(\mathbf{q})=\tfrac{1}{2}[\cos q_xa+\cos q_ya]. \tag{C4}$$

and the sum over wave vectors is over the Brillouin zone associated with the magnetic unit cell (see Fig. 7). Also

$$\mathcal{H}_\delta=\delta J_1S\sum_{\mathbf{k}}[c_x(\mathbf{k})-c_y(\mathbf{k})][a(\mathbf{k})+a^\dagger(-\mathbf{k})]\times[b^\dagger(\mathbf{k})+b(-\mathbf{k})], \tag{C5}$$

where $c_x(\mathbf{k})=\cos k_xa$ and $c_y(\mathbf{k})=\cos k_ya$.

Since the effect we wish to treat involves energies of relative order $(1/S)$, we now consider the fourth-order terms V_4 in the boson Hamiltonian, which we write as

$$V_4=V_J+V_\delta, \tag{C6}$$

where

$$V_J=-\tfrac{1}{2}J\sum_{\langle ij\rangle}b_j^\dagger(a_i^\dagger+b_j)^2a_i, \tag{C7}$$

where $\langle ij\rangle$ indicates that i is summed over a sites and j over nearest-neighboring b sites and

$$V_\delta=\delta J_1\sum_{\langle ij\rangle}\sigma_\delta[-\tfrac{1}{4}a_i^\dagger a_i^\dagger a_i(b_j^\dagger+b_j)-\tfrac{1}{4}(a_i^\dagger+a_i)b_j^\dagger b_jb_j+a_i^\dagger a_ib_j^\dagger b_j], \tag{C8}$$

where σ_δ is $+1$ for x bonds and -1 for y bonds.

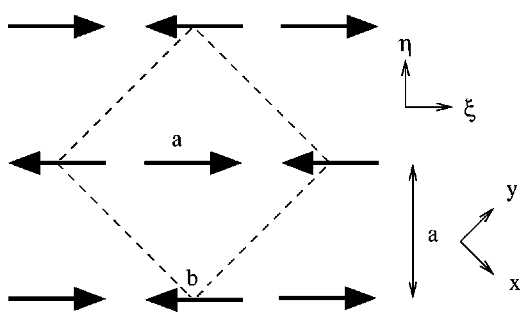

FIG. 6. Unit cell of the square lattice.

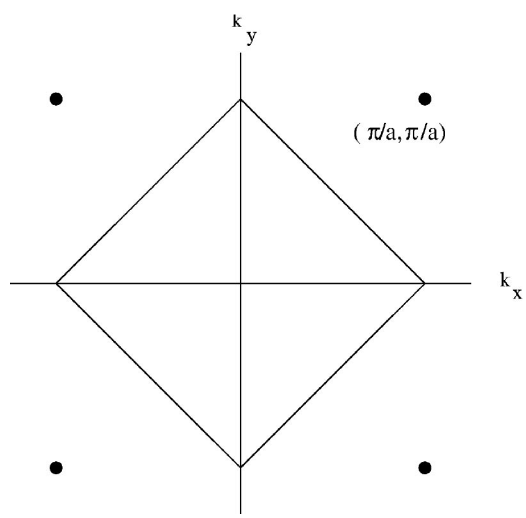

FIG. 7. Brillouin zone for the square lattice.

We construct the effective quadratic Hamiltonian by taking all possible averages of pairs of operators out of the fourth order terms. Thus we have the effective quadratic terms

$$\Delta H_J = -\tfrac{1}{2} J \sum_{\langle ij\rangle} [a_i b_j^\dagger \langle (b_j + a_i^\dagger)^2\rangle + 2 a_i (b_j + a_i^\dagger) \times \langle b_j^\dagger (b_j + a_i^\dagger)\rangle + 2 b_j^\dagger (b_j + a_i^\dagger)\langle (b_j + a_i^\dagger) a_i\rangle + (b_j + a_i^\dagger)^2 \langle a_i b_j^\dagger\rangle] \tag{C9}$$

and

$$\Delta H_\delta = \delta J_1 \sum_{\langle ij\rangle} \sigma_\delta [-\tfrac{1}{2} a_i^\dagger (b_j^\dagger + b_j)\langle a_i^\dagger a_i\rangle - \tfrac{1}{2} a_i^\dagger a_i \langle a_i^\dagger (b_j^\dagger + b_j)\rangle - \tfrac{1}{4} a_i^\dagger a_i^\dagger \langle a_i (b_j^\dagger + b_j)\rangle - \tfrac{1}{4} a_i (b_j^\dagger + b_j)\langle a_i^\dagger a_i^\dagger\rangle - \tfrac{1}{2}(a_i^\dagger + a_i) b_j \langle b_j^\dagger b_j\rangle - \tfrac{1}{2} b_j^\dagger b_j \langle b_j (a_i^\dagger + a_i)\rangle - \tfrac{1}{4}(a_i^\dagger + a_i) b_j^\dagger \langle b_j b_j\rangle - \tfrac{1}{4} b_j b_j \langle (a_i^\dagger + a_i) b_j^\dagger\rangle + a_i^\dagger a_i \langle b_j^\dagger b_j\rangle + b_j^\dagger b_j \langle a_i^\dagger a_i\rangle + a_i^\dagger b_j \langle b_j^\dagger a_i\rangle + a_i^\dagger b_j^\dagger \langle a_i b_j\rangle + b_j^\dagger a_i \langle a_i^\dagger b_j\rangle + b_j a_i \langle b_j^\dagger a_i^\dagger\rangle], \tag{C10}$$

where $\langle X\rangle$ denotes an average with respect to the quadratic Hamiltonian.

Since the quadratic Hamiltonian is real and Hermitian we can equate averages like $\langle a_i^\dagger b_j^\dagger\rangle$ and $\langle a_i b_j\rangle$. Also at this order of $(1/S)$ we only need keep Hermitian contributions to the effective Hamiltonian. Therefore we write

$$\Delta H_J = -\tfrac{1}{4} J \sum_{\langle ij\rangle} [(a_i b_j^\dagger + a_i^\dagger b_j)\langle (b_j + a_i^\dagger)^2\rangle + 4(a_i + b_j^\dagger)(b_j + a_i^\dagger)\langle b_j^\dagger (b_j + a_i^\dagger)\rangle + (b_j^\dagger + a_i)^2 \langle a_i b_j^\dagger\rangle + (b_j + a_i^\dagger)^2 \langle a_i b_j^\dagger\rangle]. \tag{C11}$$

Next we consider ΔH_δ. Here we can eliminate any terms which involve local averages (e.g., $\langle a_i^\dagger a_i\rangle$) because they multiply a function whose Fourier coefficient vanishes at zero wave vector. Thereby we have

$$\Delta H_\delta = \delta J_1 \sum_{\langle ij\rangle} \sigma_\delta [-\tfrac{1}{2} a_i^\dagger a_i \langle a_i^\dagger (b_j^\dagger + b_j)\rangle - \tfrac{1}{4} a_i^\dagger a_i^\dagger \langle a_i (b_j^\dagger + b_j)\rangle - \tfrac{1}{2} b_j^\dagger b_j \langle b_j (a_i^\dagger + a_i)\rangle - \tfrac{1}{4} b_j b_j \langle (a_i^\dagger + a_i) b_j^\dagger\rangle + a_i^\dagger b_j \langle b_j^\dagger a_i\rangle + a_i^\dagger b_j^\dagger \langle a_i b_j\rangle + b_j^\dagger a_i \langle a_i^\dagger b_j\rangle + b_j a_i \langle b_j^\dagger a_i^\dagger\rangle]. \tag{C12}$$

Taking the Hermitian part of this we get

$$\Delta H_\delta = \delta J_1 \sum_{\langle ij\rangle} \sigma_\delta [-\tfrac{1}{2} a_i^\dagger a_i \langle a_i^\dagger (b_j^\dagger + b_j)\rangle - \tfrac{1}{8}(a_i^\dagger a_i^\dagger + a_i a_i)\langle a_i (b_j^\dagger + b_j)\rangle - \tfrac{1}{2} b_j^\dagger b_j \langle b_j (a_i^\dagger + a_i)\rangle - \tfrac{1}{8}(b_j^\dagger b_j^\dagger + b_j b_j)\langle (a_i^\dagger + a_i) b_j^\dagger\rangle + (a_i^\dagger b_j + a_i b_j^\dagger)\langle b_j^\dagger a_i\rangle + (a_i^\dagger b_j^\dagger + a_i b_j)\langle a_i b_j\rangle]. \tag{C13}$$

Thus we need the averages

$$X_1 \equiv \langle b_j^2\rangle = \langle (b_j^\dagger)^2\rangle = \langle a_i^2\rangle = \langle (a_i^\dagger)^2\rangle, \tag{C14a}$$

$$X_2 \equiv \langle b_j^\dagger b_j\rangle = \langle a_i^\dagger a_i\rangle, \tag{C14b}$$

$$Y_{ij} \equiv \langle b_j a_i^\dagger\rangle = \langle b_j^\dagger a_i\rangle \equiv Y_0 + \sigma_\delta Y, \tag{C14c}$$

$$Z_{ij} \equiv \langle b_j^\dagger a_i^\dagger\rangle = \langle b_j a_i\rangle \equiv Z_0 + \sigma_\delta Z, \tag{C14d}$$

where

$$Y_0 = \tfrac{1}{4}\sum_j Y_{ij} = \tfrac{1}{4}\sum_j \langle b_j^\dagger a_i\rangle, \tag{C15a}$$

$$Y = \tfrac{1}{4}\sum_j \sigma_\delta Y_{ij} = \tfrac{1}{4}\sum_j \sigma_\delta \langle b_j^\dagger a_i\rangle, \tag{C15b}$$

$$Z_0 = \tfrac{1}{4}\sum_j Z_{ij} = \tfrac{1}{4}\sum_j \langle b_j a_i\rangle, \tag{C15c}$$

$$Z = \tfrac{1}{4}\sum_j \sigma_\delta Z_{ij} = \tfrac{1}{4}\sum_j \sigma_\delta \langle b_j a_i\rangle, \tag{C15d}$$

where the sums over j are restricted to sites that are nearest neighbors of site i. Now drop terms which sum to zero because of σ_δ and also those (such as $\Sigma_{ij}\sigma_\delta a_i^\dagger b_j$) which do not contribute at zero wave vector. Then we get

$$\Delta H_J + \Delta H_\delta = -\tfrac{1}{4} J \sum_{\langle ij\rangle} [(2X_1 + 4Y_0)(a_i b_j^\dagger + a_i^\dagger b_j) + Y_0[(b_j^\dagger)^2 + b_j^2 + a_i^2 + (a_i^\dagger)^2] + 4(X_2 + Z_0) \times (a_i^\dagger + b_j)(a_i + b_j^\dagger)] + \tfrac{1}{8}\delta J_1 \sum_{ij} [(Y+Z)[-4a_i^\dagger a_i - 4b_j^\dagger b_j - a_i^2 - (a_i^\dagger)^2 - b_j^2 - (b_j^\dagger)^2] + 8Y(a_i^\dagger b_j + a_i b_j^\dagger) + 8Z(a_i^\dagger b_j^\dagger + a_i b_j)]. \tag{C16}$$

The coefficients can be evaluated straightforwardly. For instance, if one considers $\mathcal{H}_J$ as the unperturbed Hamiltonian and treats $\mathcal{H}_\delta$ as a perturbation, then one has

$$Y=\tfrac{1}{4}\sum_j \sigma_\delta\langle b_j^\dagger a_i\rangle$$
$$=\sum_j \sigma_\delta\left[\left\langle 0\left|b_j^\dagger a_i\frac{1}{\mathcal{E}}\mathcal{H}_\delta\right|0\right\rangle+\langle 0|\mathcal{H}_\delta\frac{1}{\mathcal{E}}b_j^\dagger a_i|0\rangle\right], \tag{C17}$$

where $|0\rangle$ is the spin-wave vacuum and $\mathcal{E}$ is the unperturbed energy of the virtual state. We give the evaluations

$$X_1=4C_{2c}(\delta J_1/J)^2, \tag{C18a}$$

$$Y_0=4C_{2d}(\delta J_1/J)^2, \tag{C18b}$$

$$Y=-8C_{2a}(\delta J_1/J), \tag{C18c}$$

$$Z=-8C_{2b}(\delta J_1/J), \tag{C18d}$$

where

$$C_{2a}=\frac{1}{128N}\sum_{\mathbf{q}}\frac{[c_x(\mathbf{q})-c_y(\mathbf{q})]^2}{\epsilon(\mathbf{q})^3}, \tag{C19a}$$

$$C_{2b}=\frac{1}{128N}\sum_{\mathbf{q}}\frac{[c_x(\mathbf{q})-c_y(\mathbf{q})]^2}{\epsilon(\mathbf{q})^3}\gamma(\mathbf{q})^2, \tag{C19b}$$

$$C_{2c}=\frac{1}{128N}\sum_{\mathbf{q}}\frac{[c_x(\mathbf{q})-c_y(\mathbf{q})]^2}{\epsilon(\mathbf{q})^5}[1+2\gamma(\mathbf{q})^2], \tag{C19c}$$

$$C_{2d}=C_2-C_{2c}, \tag{C19d}$$

where $\epsilon(\mathbf{q})^2=1-\gamma(\mathbf{q})^2, C_2=C_{2a}+C_{2b}$.

To summarize, the effect of quantum fluctuations of the in-plane exchange anisotropy are contained in the effective Hamiltonian of Eq. (C16). Since the result is given in real space, we can apply it now to the 2342 structure where it gives rise to contributions to the dynamical matrices written in Eq. (69). The terms proportional to X_2+Z_0 are taken into account by the spin-wave renormalization incorporated in Z_c.

APPENDIX D: IN-PLANE ANISOTROPIC I-II INTERACTION

We start from Eq. (71), which can be written as $V_{12}=V_{12}^e+V_{12}^f$, where

$$V_{12}^e=-\delta J_{12}\sqrt{S/2}\sum_i e_i^\dagger e_i(a_{i+x}+a_{i+x}^\dagger+d_{i-x}+d_{i-x}^\dagger$$
$$-b_{i-y}^\dagger-b_{i-y}-c_{i+y}-c_{i+y}^\dagger)-4\delta J_{12}S\sqrt{S/2}$$
$$\times\sum_i\left[e_i^\dagger+e_i-\frac{e_i^\dagger e_i e_i}{2S}\right]+\delta J_{12}\sqrt{S/2}\sum_i(e_i^\dagger+e_i)$$
$$\times[a_{i+x}^\dagger a_{i+x}+d_{i-x}^\dagger d_{i-x}+b_{i-y}^\dagger b_{i-y}+c_{i+y}^\dagger c_{i+y}] \tag{D1}$$

and V_{12}^f is obtained from V_{12}^e by replacing e by $f^\dagger$, x by $-x$, y by $-y$ and normally ordering the result. So we focus on V_{12}^e. Eliminate terms linear in the boson operators by the shifts

$$e_i\rightarrow e_i+s,\quad a_i\rightarrow a_i+t,\quad b_i\rightarrow b_i+t,$$
$$c_i\rightarrow c_i+t,\quad d_i\rightarrow d_i+t. \tag{D2}$$

The corresponding Fourier transforms are shifted by a factor $\sqrt{N_{uc}}$. For example,

$$N_{\rm uc}^{-1/2}\sum_i a_i=a(0)\rightarrow a(0)+t\sqrt{N_{uc}}. \tag{D3}$$

In what follows e will denote $e(\mathbf{q}=0)$ and similarly for other operators. Then the linear terms in the Hamiltonian V_{12}^e are

$$V_1^e=-4\delta J_{12}S\sqrt{N_{uc}S/2}(e^\dagger+e). \tag{D4}$$

The quadratic zero-wave-vector terms in the isotropic part of the Hamiltonian are

$$V_2=(4J+2J_3)S(a^\dagger a+b^\dagger b+c^\dagger c+d^\dagger d)+4J_2S(e^\dagger e+f^\dagger f)$$
$$+J_{12}S([a^\dagger+d^\dagger]f+[b^\dagger+c^\dagger]e+[a+d]f^\dagger+[b+c]e^\dagger)$$
$$+(2J+2J_3)S(a^\dagger b^\dagger+c^\dagger d^\dagger+ab+cd)$$
$$+2JS(a^\dagger c^\dagger+b^\dagger d^\dagger+ac+bd)+4J_2S(e^\dagger f^\dagger+ef)$$
$$+J_{12}S([a^\dagger+d^\dagger]e^\dagger+[b^\dagger+c^\dagger]f^\dagger+[a+d]e+[b+c]f). \tag{D5}$$

We determine the shifts s and t by requiring that

$$\frac{\partial(V_1+V_2)}{\partial e}=0$$
$$=\sqrt{N_{uc}}S(-4\delta J_{12}\sqrt{S/2}+2J_{12}t$$
$$+4J_2s+2J_{12}t+4J_2s),$$

$$\frac{\partial(V_1+V_2)}{\partial a}=0$$
$$=\sqrt{N_{uc}}S[(4J+2J_3)t+J_{12}s$$
$$+(2J+2J_3)t+2Jt+J_{12}s)]. \tag{D6}$$

For $J_{12}^2\ll 4JJ_2$ we have

$$s=4\delta J_{12}\frac{\sqrt{S/2}}{8J_2}, \tag{D7}$$

$$t=-\frac{2J_{12}s}{8J+4J_3}=-J_{12}\delta J_{12}\frac{\sqrt{S/2}}{J_2(8J+4J_3)}. \tag{D8}$$

As discussed in the text, these are the expected results.

Now we record the terms in the Hamiltonian V_{12}^e which are cubic in boson operators

$$\mathcal{H}^{(3,e)}=\delta J_{12}\sqrt{S/2}\Bigg\{-\sum_{i\in e} e_i^\dagger e_i(a_{i+x}^\dagger+a_{i+x}+d_{i-x}^\dagger+d_{i-x}-b_{i-y}^\dagger-b_{i-y}-c_{i+y}^\dagger-c_{i+y})+2\sum_{i\in e} e_i^\dagger e_i e_i+\sum_{i\in e}(e_i^\dagger+e_i)(a_{i+x}^\dagger a_{i+x}+d_{i-x}^\dagger d_{i-x}+b_{i-y}^\dagger b_{i-y}+c_{i+y}^\dagger c_{i+y})\Bigg\}. \tag{D9}$$

Now make the replacements of Eq. (D2) to get the quadratic contribution from $\mathcal{H}^{(3,e)}$ as

$$\mathcal{H}^{(3,e)}=\langle e\rangle\delta J_{12}\sqrt{S/2}\Bigg\{-\sum_{i\in e}(e_i^\dagger+e_i)(a_{i+x}^\dagger+a_{i+x}+d_{i-x}^\dagger+d_{i-x}-b_{i-y}^\dagger-b_{i-y}-c_{i+y}^\dagger-c_{i+y})+2\sum_{i\in e}[e_i^2+2e_i^\dagger e_i]+2\sum_{i\in e}(a_{i+x}^\dagger a_{i+x}+d_{i-x}^\dagger d_{i-x}+b_{i-y}^\dagger b_{i-y}+c_{i+y}^\dagger c_{i+y})\Bigg\}. \tag{D10}$$

Here we dropped the terms proportional to $\langle a\rangle$. They are smaller than those in $\langle e\rangle$ by $J_{12}/(4J)\approx 1/50$. Also, as before, to this order in $1/S$ we may replace the perturbation by its Hermitian part. Then the sum of the effective quadratic terms from $\mathcal{H}^{(3,e)}$ and $\mathcal{H}^{(3,f)}$ are

$$\mathcal{H}^{(3)}=\frac{(\delta J_{12})^2S}{4J_2}\sum_{\mathbf{q}}\{4[a^\dagger(\mathbf{q})a(\mathbf{q})+b^\dagger(\mathbf{q})b(\mathbf{q})+c^\dagger(\mathbf{q})c(\mathbf{q})+d^\dagger(\mathbf{q})d(\mathbf{q})+e^\dagger(\mathbf{q})e(\mathbf{q})+f^\dagger(\mathbf{q})f(\mathbf{q})]+e^\dagger(\mathbf{q})e^\dagger(-\mathbf{q})+e(\mathbf{q})e(-\mathbf{q})+f^\dagger(\mathbf{q})f^\dagger(-\mathbf{q})+f(\mathbf{q})f(-\mathbf{q})+[e(\mathbf{q})-f(\mathbf{q})+e^\dagger(-\mathbf{q})-f^\dagger(-\mathbf{q})]\times[-a^\dagger(\mathbf{q})+b^\dagger(\mathbf{q})+c^\dagger(\mathbf{q})-d^\dagger(\mathbf{q})-a(-\mathbf{q})+b(-\mathbf{q})+c(-\mathbf{q})-d(-\mathbf{q})]\}, \tag{D11}$$

which leads to Eq. (74).

Now we look at the fourth order terms in the Cu_{II}-Cu_{II} isotropic exchange interaction. These are

$$V_{DM}=-\tfrac{1}{2}J_2\sum_{i\in e,\delta}(e_i^\dagger f_{i+\delta}^\dagger f_{i+\delta}^\dagger f_{i+\delta}+f_{i+\delta}e_i^\dagger e_i e_i+2e_i^\dagger e_i f_{i+\delta}^\dagger f_{i+\delta}). \tag{D12}$$

Substituting in two shifts of $\langle e\rangle$, this is

$$V_{DM}=-\tfrac{1}{2}\langle e\rangle^2 J_2\sum_{i,\delta}[e_i^\dagger f_{i+\delta}+2f_{i+\delta}^\dagger f_{i+\delta}+2e_i^\dagger f_{i+\delta}^\dagger+(f_{i+\delta}^\dagger)^2+2f_{i+\delta}e_i+f_{i+\delta}e_i^\dagger+2e_i^\dagger e_i+e_i^2+2e_i^\dagger e_i+2f_{i+\delta}^\dagger f_{i+\delta}+2(e_i^\dagger+e_i)(f_{i+\delta}^\dagger+f_{i+\delta})]. \tag{D13}$$

Taking the Hermitian part of this, we get

$$\begin{aligned}V_{DM}=&-\frac{(\delta J_{12})^2S}{16J_2}\sum_{i,\delta}[e_i^\dagger f_{i+\delta}+e_i f_{i+\delta}^\dagger+2e_i^\dagger f_{i+\delta}^\dagger+2e_i f_{i+\delta}+4f_{i+\delta}^\dagger f_{i+\delta}+4e_i^\dagger e_i+\tfrac{1}{2}e_i^2+\tfrac{1}{2}(e_i^\dagger)^2+\tfrac{1}{2}f_{i+\delta}^2+\tfrac{1}{2}(f_{i+\delta}^\dagger)^2+2(e_i^\dagger+e_i)(f_{i+\delta}^\dagger+f_{i+\delta})]\\ =&-\frac{(\delta J_{12})^2S}{16J_2}\sum_{\mathbf{q}}\{[6e(\mathbf{q})f^\dagger(\mathbf{q})+6e^\dagger(\mathbf{q})f(\mathbf{q})+8e(\mathbf{q})f(-\mathbf{q})+8e^\dagger(\mathbf{q})f^\dagger(-\mathbf{q})](c_x+c_y)+16f^\dagger(\mathbf{q})f(\mathbf{q})+16e^\dagger(\mathbf{q})e(\mathbf{q})+2e^\dagger(\mathbf{q})e^\dagger(-\mathbf{q})+2e(\mathbf{q})e(-\mathbf{q})+2f^\dagger(\mathbf{q})f^\dagger(-\mathbf{q})+2f(\mathbf{q})f(-\mathbf{q})\},\end{aligned} \tag{D14}$$

which leads to Eq. (75).

Contributions from quartic terms in the Cu_I-Cu_{II} interaction are smaller, i.e., of order $(\delta J_{12})^2J_{12}^2/(JJ_2^2)$, if we take out one factor of $\langle e\rangle$ and one factor of $\langle a\rangle$. Taking out two $\langle a\rangle$ factors gives an even smaller result. Taking out two $\langle a\rangle$ shifts from the Cu_I-Cu_I anharmonic term gives a contribution of order $J_{12}^2\delta J_{12}^2/(JJ_2^2)$. All these terms are neglected.

APPENDIX E: IN-PLANE ANISOTROPIC II-II INTERACTION

1. Self-energy due to cubic perturbations

We start by discussing how one constructs the self-energy due to cubic perturbations. The point is that we wish to avoid the complexities involving Matsubara sums, etc. Let us suppose that we have an unperturbed Hamiltonian in terms of normal mode operators $E(\mathbf{q})$ and $F(\mathbf{q})$:

$$\mathcal{H}=\sum_{\mathbf{q}}\omega(\mathbf{q})[E^\dagger(\mathbf{q})E(\mathbf{q})+F^\dagger(\mathbf{q})F(\mathbf{q})]. \tag{E1}$$

Now we want to identify the perturbative contributions to the matrices $\mathbf{A}(\mathbf{q})$ and $\mathbf{B}(\mathbf{q})$. Suppose we wish to calculate perturbative contributions leading to an effective quadratic Hamiltonian of the form

$$\tfrac{1}{2}B(\mathbf{q})E^\dagger(\mathbf{q})E^\dagger(-\mathbf{q}). \tag{E2}$$

For this purpose we make the identification

$$\delta B(\mathbf{q})=\left\langle 0\left|E(\mathbf{q})E(-\mathbf{q})V\frac{1}{\mathcal{E}}V\right|0\right\rangle. \tag{E3}$$

Thus for $\omega(\mathbf{q})\to 0$ and considering only the ground state, we may write

$$\delta B(\mathbf{q})=\left\langle 0\left|\partial V/\partial E^\dagger(\mathbf{q})\frac{1}{\mathcal{E}}\partial V/\partial E^\dagger(-\mathbf{q})\right|0\right\rangle+\left\langle 0\left|\partial V/\partial E^\dagger(-\mathbf{q})\frac{1}{\mathcal{E}}\partial V/\partial E^\dagger(\mathbf{q})\right|0\right\rangle. \tag{E4}$$

Similarly for the term in the Hamiltonian

$$A(\mathbf{q})E^\dagger(\mathbf{q})F(\mathbf{q}) \tag{E5}$$

we make the identification

$$\delta A(\mathbf{q}) = \left\langle 0 \left| E(\mathbf{q}) V \frac{1}{\mathcal{E}} V F^\dagger(\mathbf{q}) \right| 0 \right\rangle. \tag{E6}$$

Thus for $\omega(\mathbf{q}) \to 0$ and considering only the ground state, we may write

$$\delta A(\mathbf{q}) = \left\langle 0 \left| \partial V/\partial E^\dagger(\mathbf{q}) \frac{1}{\mathcal{E}} \partial V/\partial F(\mathbf{q}) \right| 0 \right\rangle + \left\langle 0 \left| \partial V/\partial F(\mathbf{q}) \frac{1}{\mathcal{E}} \partial V/\partial E^\dagger(\mathbf{q}) \right| 0 \right\rangle. \tag{E7}$$

This type of relation holds generally under the two assumptions: (a) we consider the perturbation to modes whose energy can be neglected in the energy denominators and (b) we consider only the ground and low lying excited states, so that boson occupation numbers are zero. We have made the identification in terms of the normal mode operators, but equally we may transform to any set of modes.

2. Application to Cu_{II}-Cu_{II} in-plane interactions

We start from Eq. (76) and implement the results of the preceding subsection. For small $\mathbf{k}$ we write

$$\begin{aligned}
T_1 &\equiv \partial V/\partial e(\mathbf{k}) \\
&= \delta J_2 \sqrt{2S/N_{\rm uc}} \sum_{\mathbf{q}} [f(\mathbf{q}) + f^\dagger(-\mathbf{q})] e^\dagger(\mathbf{q})(c_x - c_y), \\
T_2 &\equiv \partial V/\partial e^\dagger(\mathbf{k}) \\
&= \delta J_2 \sqrt{2S/N_{\rm uc}} \sum_{\mathbf{q}} [f(\mathbf{q}) + f^\dagger(-\mathbf{q})] e(-\mathbf{q})(c_x - c_y), \\
T_3 &\equiv \partial V/\partial f(\mathbf{k}) \\
&= -\delta J_2 \sqrt{2S/N_{\rm uc}} \sum_{\mathbf{q}} [e(\mathbf{q}) + e^\dagger(-\mathbf{q})] f^\dagger(\mathbf{q})(c_x - c_y), \\
T_4 &\equiv \partial V/\partial f^\dagger(\mathbf{k}) \\
&= -\delta J_2 \sqrt{2S/N_{\rm uc}} \sum_{\mathbf{q}} [e(\mathbf{q}) + e^\dagger(-\mathbf{q})] f(-\mathbf{q})(c_x - c_y),
\end{aligned} \tag{E8}$$

where $c_x = \cos(aq_x)$ and $c_y = \cos(aq_y)$. Thus if $\overline{\mathbf{p}}$ denotes $-\mathbf{p}$, then

$$\begin{aligned}
\left\langle T_1 \frac{1}{\mathcal{E}} T_1 \right\rangle &= \frac{2S(\delta J_2)^2}{N_{\rm uc}} \sum_{\mathbf{q},\mathbf{p}} \left\langle [f(\mathbf{q}) + f^\dagger(\overline{\mathbf{q}})] e^\dagger(\mathbf{q})(c_x - c_y) \times \frac{1}{\mathcal{E}} [f(\mathbf{p}) + f^\dagger(\overline{\mathbf{p}})] e^\dagger(\mathbf{p})(c_x - c_y) \right\rangle \\
&= \frac{2S(\delta J_2)^2}{N_{\rm uc}} \sum_{\mathbf{q},\mathbf{p}} \left\langle [l_{\mathbf{q}} \delta_{\mathbf{q}} - m_{\mathbf{q}} \eta_{\mathbf{q}}](-m_{\mathbf{q}} \delta_{\overline{\mathbf{q}}}) (c_x - c_y) \frac{1}{\mathcal{E}} [-m_{\mathbf{p}} \eta^\dagger_{\mathbf{p}} + l_{\mathbf{p}} \delta^\dagger_{\overline{\mathbf{p}}}] l_{\mathbf{p}} \eta^\dagger_{\mathbf{p}} (c_x - c_y) \right\rangle \\
&= -\frac{2S(\delta J_2)^2}{N_{\rm uc}} \sum_{\mathbf{q}} \frac{(c_x - c_y)^2 l_{\mathbf{q}}^2 m_{\mathbf{q}}^2}{8 J_2 S \epsilon(\mathbf{q})} \\
&= -\frac{(\delta J_2)^2}{4 J_2 N_{\rm uc}} \sum_{\mathbf{q}} \left(\frac{(c_x - c_y)^2}{\epsilon(\mathbf{q})^3} \right) \frac{\gamma(\mathbf{q})^2}{4},
\end{aligned} \tag{E9}$$

where the normal mode operators $\eta(\mathbf{q})$ and $\delta(\mathbf{q})$ were introduced in Eq. (B9). Similarly

$$\begin{aligned}
\left\langle T_1 \frac{1}{\mathcal{E}} T_2 \right\rangle = \left\langle T_2 \frac{1}{\mathcal{E}} T_1 \right\rangle
&= -\frac{(\delta J_2)^2}{4 J_2 N_{\rm uc}} \sum_{\mathbf{q}} \left(\frac{(c_x - c_y)^2}{\epsilon(\mathbf{q})^3} \right) \times \left(\frac{1 - \epsilon(\mathbf{q})}{2} + \frac{\gamma(\mathbf{q})^2}{4} \right),
\end{aligned} \tag{E10}$$

$$\left\langle T_1 \frac{1}{\mathcal{E}} T_3 \right\rangle = \left\langle T_3 \frac{1}{\mathcal{E}} T_1 \right\rangle = \frac{(\delta J_2)^2}{4 J_2 N_{\rm uc}} \sum_{\mathbf{q}} \left(\frac{(c_x - c_y)^2}{\epsilon(\mathbf{q})^3} \right) \frac{3\gamma(\mathbf{q})^2}{4}, \tag{E11}$$

$$\left\langle T_1 \frac{1}{\mathcal{E}} T_4 \right\rangle = \left\langle T_4 \frac{1}{\mathcal{E}} T_1 \right\rangle = \frac{(\delta J_2)^2}{4 J_2 N_{\rm uc}} \sum_{\mathbf{q}} \left(\frac{(c_x - c_y)^2}{\epsilon(\mathbf{q})^3} \right) \frac{[1 - \epsilon(\mathbf{q})]^2}{4}. \tag{E12}$$

Now we have the contribution to the coefficient of $e^\dagger e$, which we denote δa_{55}, as

$$\delta a_{55} = \left\langle T_1 \frac{1}{\mathcal{E}} T_2 \right\rangle + \left\langle T_2 \frac{1}{\mathcal{E}} T_1 \right\rangle = [\delta J_2^2/J_2][-32 C_{2a} - 16 C_{2b}], \tag{E13}$$

where C_{2a} and C_{2b} were defined in Eq. (C19).

Likewise the contribution to the coefficient of $e^\dagger f$ which we denote δa_{56} is

$$\delta a_{56} = \left\langle T_1 \frac{1}{\mathcal{E}} T_4 \right\rangle + \left\langle T_4 \frac{1}{\mathcal{E}} T_1 \right\rangle = [(\delta J_2)^2/J_2][32 C_{2a} - 16 C_{2b}]. \tag{E14}$$

Similarly, δb_5 is the contribution to the coefficient of $\frac{1}{2} e^\dagger e^\dagger$, so that

$$\delta b_{55} = 2 \left\langle T_1 \frac{1}{\mathcal{E}} T_1 \right\rangle = [(\delta J_2)^2/J_2][-16 C_{2b}] \tag{E15}$$

and likewise

$$\delta b_{56}=\left\langle T_1\frac{1}{\mathcal{E}}T_3\right\rangle+\left\langle T_3\frac{1}{\mathcal{E}}T_1\right\rangle=[(\delta J_2)^2/J_2][48C_{2b}]. \tag{E16}$$

APPENDIX F: INTERPLANAR ANISOTROPIC Cu_{II}-Cu_{II} INTERACTION

1. Pseudodipolar interactions

In order to facilitate the evaluation of the lattice sums we parametrize the anisotropic exchange interactions between the ith Cu_{II} spin in one plane and the nearest-neighboring jth Cu_{II} spin in an adjacent layer. We introduce the indicator variable σ_i which is unity if i is on the e sublattice and is -1 if i is on the f sublattice. We also introduce a variable μ_i to distinguish between the two nearest-neighboring sites with the same value of σ_i. Then for the interaction between nearest-neighboring Cu_{II} spins i and j in adjacent CuO layers we use Fig. 4 to write the principal axes as

$$\hat{n}_1^{(ij)}=[\tfrac{1}{2}(1+\sigma_i\sigma_j)\hat{\eta}-\tfrac{1}{2}(1-\sigma_i\sigma_j)\hat{\xi}]\mu_i\mu_j, \tag{F1a}$$

$$\hat{n}_2^{(ij)}=[\tfrac{1}{2}(1+\sigma_i\sigma_j)\hat{\xi}\cos\psi+\tfrac{1}{2}(1-\sigma_i\sigma_j)\hat{\eta}\cos\psi]\mu_i\mu_j+\hat{z}\sin\psi, \tag{F1b}$$

$$\hat{n}_3^{(ij)}=[\tfrac{1}{2}(1+\sigma_i\sigma_j)\hat{\xi}\sin\psi+\tfrac{1}{2}(1-\sigma_i\sigma_j)\hat{\eta}\sin\psi]\mu_i\mu_j-\hat{z}\cos\psi. \tag{F1c}$$

We also write

$$\mathbf{S}_i=-\sigma_i(S-a_i^\dagger a_i)\hat{\xi}+\sqrt{S/2}(a_i^\dagger+a_i)\hat{\eta}+i\sqrt{S/2}(a_i-a_i^\dagger)\hat{z}\sigma_i, \tag{F2}$$

where, in this appendix, a_i is the boson operator for spin i. Then we have

$$\mathbf{S}_i\cdot\hat{n}_1^{(ij)}=\tfrac{1}{2}(\sigma_i-\sigma_j)\mu_i\mu_j(S-a_i^\dagger a_i)+\tfrac{1}{2}(1+\sigma_i\sigma_j)\times\mu_i\mu_j\sqrt{S/2}(a_i^\dagger+a_i), \tag{F3a}$$

$$\mathbf{S}_i\cdot\hat{n}_2^{(ij)}=-\tfrac{1}{2}(S-a_i^\dagger a_i)c(\sigma_i+\sigma_j)\mu_i\mu_j+\tfrac{1}{2}(1-\sigma_i\sigma_j)\times\mu_i\mu_j c\sqrt{S/2}(a_i^\dagger+a_i)+is\sigma_i\sqrt{S/2}(a_i-a_i^\dagger), \tag{F3b}$$

$$\mathbf{S}_i\cdot\hat{n}_3^{(ij)}=-\tfrac{1}{2}(S-a_i^\dagger a_i s(\sigma_i+\sigma_j)\mu_i\mu_j+\tfrac{1}{2}(1-\sigma_i\sigma_j)\times\mu_i\mu_j s\sqrt{S/2}(a_i^\dagger+a_i)-ic\sigma_i\sqrt{S/2}(a_i-a_i^\dagger), \tag{F3c}$$

where $c\equiv\cos\psi$ and $s\equiv\sin\psi$. Then we have

$$\mathcal{H}_{ij}=S\sum_{m=1}^{3}K_m[\mathbf{S}_i\cdot\hat{n}_m][\mathbf{S}_j\cdot\hat{n}_m]\equiv S\sum_{m=1}^{3}K_mT_m, \tag{F4}$$

where, at quadratic order

$$T_1=\tfrac{1}{2}(1-\sigma_i\sigma_j)(a_i^\dagger a_i+a_j^\dagger a_j)+\tfrac{1}{4}(1+\sigma_i\sigma_j)(a_i^\dagger+a_i)(a_j^\dagger+a_j)$$

$$T_2=-\tfrac{1}{2}(1+\sigma_i\sigma_j)c^2(a_i^\dagger a_i+a_j^\dagger a_j)+\tfrac{1}{2}[\tfrac{1}{2}(1-\sigma_i\sigma_j)(a_i^\dagger+a_i)c\mu_i\mu_j+i\sigma_i s(a_i-a_i^\dagger)]\times[\tfrac{1}{2}(1-\sigma_i\sigma_j)(a_j^\dagger+a_j)c\mu_i\mu_j+i\sigma_j s(a_j-a_j^\dagger)] \tag{F5}$$

and T_3 is obtained from T_2 by replacing $\sin\psi$ by $-\cos\psi$ and $\cos\psi$ by $\sin\psi$. Thereby we get the site-diagonal contribution to the Hamiltonian as

$$\delta\mathcal{H}=4\Delta KS\sum_i a_i^\dagger a_i, \tag{F6}$$

where ΔK was defined in Eq. (83).

The remaining contributions to the Hamiltonian are found from Eq. (F4) to be

$$\delta\mathcal{H}=\tfrac{1}{2}\sum_{i\in\mathrm{II},j}\{\tfrac{1}{4}K_1S(1+\sigma_i\sigma_j)(a_i^\dagger+a_i)(a_j^\dagger+a_j)+\tfrac{1}{2}K_2S[\tfrac{1}{2}c^2(1-\sigma_i\sigma_j)(a_i^\dagger+a_i)(a_j^\dagger+a_j)-\sigma_i\sigma_j s^2(a_i-a_i^\dagger)(a_j-a_j^\dagger)+ics\mu_i\mu_j(\sigma_j-\sigma_i)\times(a_i^++a_i)(a_j-a_j^\dagger)]+\cdots\}, \tag{F7}$$

where $\cdots$ indicates further terms in K_3 obtained from those of K_2 by replacing $\cos\psi$ by $\sin\psi$ and $\sin\psi$ by $-\cos\psi$ and j is summed over Cu_{II} nearest neighbors in adjacent planes. For $q_x=q_y=0$ the imaginary term gives zero contribution to the dynamical matrices. Then, the terms with $\sigma_i=\sigma_j$ give a contribution to the Hamiltonian of

$$\delta\mathcal{H}=\frac{S}{4}\sum_i\sum_{j:\sigma_j=\sigma_i}[K_1(a_i^\dagger+a_i)(a_j^\dagger+a_j)-K_2s^2(a_i-a_i^\dagger)\times(a_j-a_j^\dagger)-K_3c^2(a_i-a_i^\dagger)(a_j-a_j^\dagger)]. \tag{F8}$$

The terms with $\sigma_i=-\sigma_j$ give a contribution to the Hamiltonian of

$$\delta\mathcal{H}=\frac{S}{4}\sum_i\sum_{j:\sigma_j=-\sigma_i}[K_2c^2(a_i+a_i^\dagger)(a_j+a_j^\dagger)+K_2s^2(a_i-a_i^\dagger)(a_j-a_j^\dagger)+K_3s^2(a_i+a_i^\dagger)(a_j+a_j^\dagger)+K_3c^2(a_i-a_i^\dagger)(a_j-a_j^\dagger)]. \tag{F9}$$

The term in Eq. (F6) and the number conserving terms in Eq. (F8) reproduce Eq. (82a) and the other terms in Eq. (F8) reproduce Eq. (82b). Equation (F9) reproduces Eqs. (82b) and (82c).

2. Dipolar interactions

For the dipolar interactions it is convenient to construct the Hamiltonian explicitly rather than to identify it with the pseudodipolar interaction. We substitute Eq. (F2) into the dipolar interaction to get

$$\begin{aligned}\mathcal{H}_{ij}&=g^2\mu_B^2 r_{ij}^{-3}[\mathbf{S}_i\cdot\mathbf{S}_j-3(\mathbf{S}_i\cdot\hat{\mathbf{r}}_{ij})(\mathbf{S}_j\cdot\hat{\mathbf{r}}_{ij})]\\&\rightarrow-3g^2\mu_B^2 r_{ij}^{-3}(\mathbf{S}_i\cdot\hat{\mathbf{r}}_{ij})(\mathbf{S}_j\cdot\hat{\mathbf{r}}_{ij})\\&=-\frac{3g^2\mu_B^2}{r_{ij}^3}\{-\sigma_i[S-a_i^\dagger a_i](\hat{\xi}\cdot\hat{\mathbf{r}}_{ij})+\sqrt{S/2}(a_i+a_i^\dagger)\\&\quad\times(\hat{\eta}\cdot\hat{\mathbf{r}}_{ij})+i\sigma_i\sqrt{S/2}(a_i-a_i^\dagger)(\hat{z}\cdot\mathbf{r}_{ij})\}\\&\quad\times\{-\sigma_j[S-a_j^\dagger a_j](\hat{\xi}\cdot\hat{\mathbf{r}}_{ij})+\sqrt{S/2}(a_j+a_j^\dagger)\\&\quad\times(\hat{\eta}\cdot\hat{\mathbf{r}}_{ij})+i\sigma_j\sqrt{S/2}(a_j-a_j^\dagger)(\hat{z}\cdot\mathbf{r}_{ij})\}.\end{aligned}\qquad\text{(F10)}$$

Here we dropped the term in $\mathbf{S}_i\cdot\mathbf{S}_j$ which may be included in the isotropic Heisenberg Hamiltonian. At quadratic order this gives

$$\begin{aligned}\mathcal{H}&=\tfrac{1}{2}\sum_{i,j\in\mathrm{II}}\mathcal{H}_{ij}\\&=\sum_{i,j\in\mathrm{II}}\frac{3g^2\mu_B^2S}{2r_{ij}^3}[\sigma_i\sigma_j(a_j^\dagger a_j+a_i^\dagger a_i)(\hat{\xi}\cdot\hat{\mathbf{r}}_{ij})^2-\tfrac{1}{2}(a_i+a_i^\dagger)\\&\quad\times(a_j+a_j^\dagger)(\hat{\eta}\cdot\mathbf{r}_{ij})^2+\tfrac{1}{2}\sigma_i\sigma_j(a_i-a_i^\dagger)(a_j-a_j^\dagger)(\hat{z}\cdot\mathbf{r}_{ij})^2\\&\quad-i\sigma_j(a_i+a_i^\dagger)(a_j-a_j^\dagger)(\hat{z}\cdot\hat{\mathbf{r}}_{ij})(\hat{\eta}\cdot\hat{\mathbf{r}}_{ij})].\end{aligned}\qquad\text{(F11)}$$

We now consider what contributions this gives to the dynamical matrix for $q_x=q_y=0$. Then the imaginary term can be dropped. For simplicity we truncate the sums to include only interactions between adjacent planes. Then we have

$$\begin{aligned}\delta a_{55}&=\sum_{j\in e}\frac{3g^2\mu_B^2S}{r_{ij}^3}[(\hat{\xi}\cdot\hat{r}_{ij})^2-\tfrac{1}{2}c_z(\hat{\eta}\cdot\hat{r}_{ij})^2-\tfrac{1}{2}c_z(\hat{z}\cdot\hat{r}_{ij})^2]\\&\quad+\sum_{j\in f}\frac{3g^2\mu_B^2S}{r_{ij}^3}\sigma_j(\hat{\xi}\cdot\hat{r}_{ij})^2,\end{aligned}\qquad\text{(F12a)}$$

$$\delta a_{56}=\sum_{j\in f}\frac{3g^2\mu_B^2S}{r_{ij}^3}[-\tfrac{1}{2}(\hat{\eta}\cdot\hat{r}_{ij})^2+\tfrac{1}{2}(\hat{z}\cdot\hat{r}_{ij})^2]c_z,\qquad\text{(F12b)}$$

$$\delta b_{55}=\sum_{j\in e}\frac{3g^2\mu_B^2S}{r_{ij}^3}[-\tfrac{1}{2}(\hat{\eta}\cdot\hat{r}_{ij})^2+\tfrac{1}{2}(\hat{z}\cdot\hat{r}_{ij})^2]c_z,\qquad\text{(F12c)}$$

$$\delta b_{56}=\sum_{j\in f}\frac{3g^2\mu_B^2S}{r_{ij}^3}[-\tfrac{1}{2}(\hat{\eta}\cdot\hat{r}_{ij})^2-\tfrac{1}{2}(\hat{z}\cdot\hat{r}_{ij})^2]c_z,\qquad\text{(F12d)}$$

where $c_z=\cos(q_zc/2)$, i is a fixed site in the e sublattice, and the sum over j is restricted to the planes adjacent to site i.

This interaction is negligibly small except with respect to the lowest in-plane mode. So we only need the combination

$$\begin{aligned}\delta&(a_{55}+b_{55}-a_{56}-b_{56})\\&=\sum_{j\in\mathrm{II}:z_{ij}=\pm c/2}\frac{3g^2\mu_B^2S}{r_{ij}^3}[\sigma_j(\hat{\xi}\cdot\hat{r}_{ij})^2-\sigma_j(\hat{\eta}\cdot\hat{r}_{ij})^2c_z]\\&=\sum_{j\in\mathrm{II}:z_{ij}=c/2}\frac{3g^2\mu_B^2S}{r_{ij}^5}[\sigma_j(x_{ij}+y_{ij})^2-\sigma_jc_z(x_{ij}-y_{ij})^2].\end{aligned}\qquad\text{(F13)}$$

Note that the sum over sites j in an adjacent plane from site i vanishes:

$$\sum_{j\in\mathrm{II}:z_{ij}=c/2}\frac{\sigma_jx_{ij}^2}{r_{ij}^5}=\sum_{j\in II:z_{ij}=c/2}\frac{\sigma_jy_{ij}^2}{r_{ij}^5}=0.\qquad\text{(F14)}$$

Thus

$$\delta(a_{55}+b_{55}-a_{56}-b_{56})=6(1+c_z)g^2\mu_B^2S\sum_{j\in II:z_{ij}=c/2}\frac{\sigma_jx_{ij}y_{ij}}{r_{ij}^5}.\qquad\text{(F15)}$$

APPENDIX G: INTERPLANAR ANISOTROPIC Cu_I-Cu_{II} INTERACTION

For the Cu_I sites we introduce further indicator variables τ (which tells the direction of the moment) and ρ (which discriminates between sublattices) such that $\tau=\rho=1$ for an a site, $-\tau=\rho=1$ for a b site, $\tau=\rho=-1$ for a c site, and $\tau=-\rho=1$ for a d site. Then, from Fig. 5, we have the principal axes for the sites i and j where i (j) is in the Cu_I (Cu_{II}) sublattice as

$$\hat{m}_1^{(ij)}=-\frac{\rho_i\sigma_j}{\sqrt{2}}[\hat{\xi}+\tau_i\hat{\eta}],\qquad\text{(G1a)}$$

$$\hat{m}_2^{(ij)}=\hat{z}\cos\phi+\frac{\rho_i\sigma_j\sin\phi}{\sqrt{2}}[\hat{\eta}-\tau_i\hat{\xi}],\qquad\text{(G1b)}$$

$$\hat{m}_3^{(ij)}=-\hat{z}\sin\phi+\frac{\rho_i\sigma_j\cos\phi}{\sqrt{2}}[\hat{\eta}-\tau_i\hat{\xi}].\qquad\text{(G1c)}$$

In checking the above it is useful to note that changing the sign of either ρ_i or σ_j induces a 180° rotation about the z axis.

Also we use Eq. (F2) for the Cu_{II} spins and

$$\mathbf{S}_i=\tau_i(S-a_i^\dagger a_i)\hat{\xi}+\sqrt{S/2}(a_i+a_i^\dagger)\hat{\eta}+i\tau_i\sqrt{S/2}(a_i^\dagger-a_i)\hat{z}\qquad\text{(G2)}$$

for the Cu_I spins. Thus if i labels a Cu_I spin we have

$$\hat{m}_1^{(ij)}\cdot \mathbf{S}_i=\frac{\rho_i\tau_i\sigma_j}{\sqrt{2}}[-(S-a_i^\dagger a_i)-\sqrt{S/2}(a_i+a_i^\dagger)], \tag{G3a}$$

$$\hat{m}_2^{(ij)}\cdot \mathbf{S}_i=\frac{1}{\sqrt{2}}[-\rho_i\sigma_j s(S-a_i^\dagger a_i)+\rho_i\sigma_j s\sqrt{S/2}(a_i+a_i^\dagger)+i\tau_i c\sqrt{S}(a_i^\dagger-a_i)], \tag{G3b}$$

$$\hat{m}_3^{(ij)}\cdot \mathbf{S}_i=\frac{1}{\sqrt{2}}[-\rho_i\sigma_j c(S-a_i^\dagger a_i)+\rho_i\sigma_j c\sqrt{S/2}(a_i+a_i^\dagger)-i\tau_i s\sqrt{S}(a_i^\dagger-a_i)] \tag{G3c}$$

and if j labels a Cu_{II} spin we have

$$\hat{m}_1^{(ij)}\cdot \mathbf{S}_j=\frac{\rho_i}{\sqrt{2}}[(S-a_j^\dagger a_j)-\tau_i\sigma_j\sqrt{S/2}(a_j+a_j^\dagger)], \tag{G4a}$$

$$\hat{m}_2^{(ij)}\cdot \mathbf{S}_j=\frac{1}{\sqrt{2}}[\rho_i\tau_i s(S-a_j^\dagger a_j)+\rho_i\sigma_j s\sqrt{S/2}(a_j+a_j^\dagger)+i\sigma_j c\sqrt{S}(a_j-a_j^\dagger)], \tag{G4b}$$

$$\hat{m}_3^{(ij)}\cdot \mathbf{S}_j=\frac{1}{\sqrt{2}}[\rho_i\tau_i c(S-a_j^\dagger a_j)+\rho_i\sigma_j c\sqrt{S/2}(a_j+a_j^\dagger)-i\sigma_j s\sqrt{S}(a_j-a_j^\dagger)],, \tag{G4c}$$

where $c\equiv\cos\phi$ and $s\equiv\sin\phi$. We now write

$$\mathcal{H}_{ij}=\sum_{m=1}^{3}[\hat{m}_m\cdot\mathbf{S}_i]\sum_{m=1}^{3}[\hat{m}_m\cdot\mathbf{S}_j]\equiv S\sum_{m=1}^{3}K_m'T_m, \tag{G5}$$

and at quadratic order we have

$$T_1=\tfrac{1}{2}\tau_i\sigma_j[a_i^\dagger a_i+a_j^\dagger a_j]+\tfrac{1}{4}[a_i+a_i^\dagger][a_j+a_j^\dagger], \tag{G6a}$$

$$T_2=\tfrac{1}{2}\tau_i\sigma_j s^2[a_i^\dagger a_i+a_j^\dagger a_j]+\left[\tfrac{1}{2}\rho_i\sigma_j s(a_i+a_i^\dagger)+\frac{i\tau_i c}{\sqrt{2}}(a_i^\dagger-a_i)\right]\times\left[\tfrac{1}{2}\rho_i\sigma_j s(a_j+a_j^\dagger)+\frac{i\sigma_j c}{\sqrt{2}}(a_j-a_j^\dagger)\right], \tag{G6b}$$

$$T_3=\tfrac{1}{2}\tau_i\sigma_j c^2[a_i^\dagger a_i+a_j^\dagger a_j]\left[\tfrac{1}{2}\rho_i\sigma_j c(a_i+a_i^\dagger)-\frac{i\tau_i s}{\sqrt{2}}(a_i^\dagger-a_i)\right]+\left[\tfrac{1}{2}\rho_i\sigma_j c(a_j+a_j^\dagger)-\frac{i\sigma_j s}{\sqrt{2}}(a_j-a_j^\dagger)\right]. \tag{G6c}$$

We drop terms which do not contribute to the dynamical matrix for $q_x=q_y=0$ and thereby find that

$$\mathcal{H}=\sum_{i\in \mathrm{II}, j\in \mathrm{II}}\mathcal{H}_{ij}=S\sum_{i\in \mathrm{I}, j\in \mathrm{II}}\{\tfrac{1}{4}(a_i^\dagger+a_i)(a_j^\dagger+a_j)(K_1'+K_2's^2+K_3'c^2)-\tfrac{1}{2}(a_i^\dagger-a_i)(a_j-a_j^\dagger)(K_2'c^2+K_3's^2)\tau_i\sigma_j\}. \tag{G7}$$

This result reproduces that of Eq. (90).

[1] R. J. Birgeneau and G. Shirane, in *Physical Properties of High Temperature Superconductors*, edited by D. M. Ginzberg (World Scientific, Singapore, 1989).
[2] M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. Endoh, Rev. Mod. Phys. **70**, 897 (1998).
[3] F. C. Chou, D. Kleinberg, M. A. Kastner R. J. Birgeneau, M. Greven, Y. J. Kim, Y. S. Lee, Amnon Aharony, O. Entin-Wohlman, A. B. Harris, and Q. Zhu, Phys. Rev. Lett. **78**, 535 (1997).
[4] M. A. Kastner, A. Aharony, R. J. Birgeneau, F. C. Chou, O. Entin-Wohlman, M. Greven, A. B. Harris, Y. J. Kim, Y. S. Lee, M. E. Parks, and Q. Zhu, Phys. Rev. B **59**, 14 702 (1999).
[5] Y. J. Kim, A. Aharony, R. J. Birgeneau, F. C. Chou, O. Entin-Wohlman, R. W. Erwin, M. Greven, A. B. Harris, M. A. Kastner, I. Ya. Korenblit, Y. S. Lee, and G. Shirane, Phys. Rev. Lett. **83**, 852 (1999).
[6] B. Grande and H. Müller-Buschbaum, Z. Naturforsch. B **31**, 405 (1976); H. Müller-Buschbaum, Angew. Chem. Int. Ed. Engl. **16**, 674 (1977).
[7] Y. J. Kim, R. J. Birgeneau, F. C. Chou, M. Greven, M. A. Kastner, Y. S. Lee, B. O. Wells, A. Aharony, O. Entin-Wolhman, I. Ya. Korenblit, A. B. Harris, R. W. Erwin, and G. Shirane, preceding paper, Phys. Rev. B **64**, 024435 (2001).
[8] E. F. Shender, Zh. Eksp. Teor. Fiz. **83**, 326 (1982) [Sov. Phys. JETP **56**, 178 (1982)].
[9] A. G. Gukasov, T. Brückel, B. Dorner, V. P. Plakhty, W. Prandl, E. F. Shender, and O. P. Smirnov, Europhys. Lett. **7**, 83 (1988); T. Bruckel, B. Dorner, A. G. Gukasov, V. P. Plakhty, W. Prandl, E. F. Shender, and O. P. Smirnow, Z. Phys. B: Condens. Matter **72**, 477 (1988); T. Brueckel, B. Dorner, A. Gukasov, and V. Plakhty, Phys. Lett. A **162**, 357 (1992).
[10] T. Yildirim, A. B. Harris, O. Entin-Wohlman, and Amnon Aharony, Phys. Rev. Lett. **72**, 3710 (1994).
[11] F. Keffer, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1966), Vol. 18/2, p. 1.
[12] T. Yildirim, A. B. Harris, O. Entin-Wohlman, and Amnon Aharony, Phys. Rev. Lett. **73**, 2919 (1994).
[13] T. Yildirim, A. B. Harris, Amnon Aharony, and O. Entin-Wohlman, Phys. Rev. B **52**, 10 239 (1995).
[14] O. Entin-Wohlman, A. B. Harris, and Amnon Aharony, Phys. Rev. B **53**, 11 661 (1996).
[15] M. Hagiwara, K. Katsumata, J. Satooka, and M. Tokunaga (unpublished).
[16] K. Katsumata, M. Hagiwara, Z. Honda, J. Satooka, A. Aharony,

O. Entin-Wohlman, R. J. Birgeneau, F. C. Chou, M. A. Kastner, Y. J. Kim, Y. S. Lee, and A. B. Harris, Europhys. Lett. (to be published).

[17] F. J. Dyson, Phys. Rev. **102**, 1217 (1956); **102**, 1230 (1956); S. V. Maleev, Zh. Eksp. Teor. Fiz. **33**, 1010 (1958) [Sov. Phys. JETP **6**, 776 (1958)].

[18] E. Rastelli and A. Tassi, J. Phys.: Condens. Matter **4**, 1567 (1992).

[19] Ravi Sachidanandam, T. Yildirim, A. B. Harris, Amnon Aharony, and O. Entin-Wohlman, Phys. Rev. B **56**, 260 (1997).

[20] R. Coldea, S. M. Hayden, G. Aeppli, T. G. Perring, C. D. Frost, T. E. Mason, S.-W. Cheong, and Z. Fisk, Physica B **276-278**, 592 (2000), show that at high energies the spin-wave modes are significantly affected by ring exchange involving four sites. We have not included this interaction in our analysis. At low energy such an interaction would probably amount to a change in J of about 5%.

[21] B. B. Beard, R. J. Birgeneau, M. Greven, and U. J. Wiese, Phys. Rev. Lett. **80**, 1742 (1998).

[22] T. Yildirim, A. B. Harris, and E. F. Shender, Phys. Rev. B **53**, 6455 (1996).

[23] T. Yildirim, A. B. Harris, and E. F. Shender, Phys. Rev. B **58**, 3144 (1998).

[24] E. Rastelli and A. Tassi, J. Phys. C **20**, L303 (1987); C. L. Henley, Phys. Rev. Lett. **62**, 2056 (1989); **73**, 2788 (1994).

[25] D. Petitgrand, S. V. Maleyev, Ph. Bourges, and A. S. Ivanov, Phys. Rev. B **59**, 1079 (1999).

[26] S. Tornow, O. Entin-Wohlman, and A. Aharony, Phys. Rev. B **60**, 10 206 (1999).

[27] V. Yu. Yushankhai and R. Hayn, cond-mat/9902106 (unpublished).

[28] I. P. R. Moreira, F. Illas, C. J. Calzado, J. F. Sanz, J.-P. Malrieu, N. B. Amor, and D. Maynau, Phys. Rev. B **59**, R6593 (1999).

[29] B. I. Halperin and P. C. Hohenberg, Phys. Rev. **117**, 952 (1969).

[30] Y. J. Kim, Ph.D. thesis, Harvard University, 2000, used the data for LCO in Ref. 31 and for 2122 in Ref. 32, to estimate J via an interpolation with respect to lattice constant.

[31] S. M. Hayden, G. Aeppli, R. Osborn, A. D. Taylor, T. G. Perring, S. W. Cheong, and Z. Fisk, Phys. Rev. Lett. **67**, 3622 (1991).

[32] M. Greven, R. J. Birgeneau, Y. Endoh, M. A. Kastner, M. Matsuda, and G. Shirane, Z. Phys. B: Condens. Matter **96**, 465 (1995).

[33] R. R. P. Singh and M. P. Gelfand, Phys. Rev. B **52**, 15 695 (1995).

[34] F. C. Chou (private communication).

[35] D. N. Zubarev, Ups. Fiz. Nauk **71**, 71 (1960) [Sov. Phys. Usp. **3**, 320 (1960)].