Selection of the ground state in type-I fcc antiferromagnets in an external magnetic field

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Selection of the ground-state spin structure through quantum fluctuations is investigated in type-I fcc antiferromagnets. Second-order real-space perturbation theory is used to account for the quantum effects in the most strongly coupled spin-spin pairs. We find that the ground state for isotropic Heisenberg spin-spin interactions is a single-**k** state in fields below $B = 0.407B_c$, where a discontinuous transition takes place to a triple-**k** structure, stable up to the transition at $B = B_c$ to the fully polarized state. The triple-**k** structure assumes a particularly simple, up-up-up-down configuration at $B = 0.5B_c$. We also study type-I fcc antiferromagnets with easy-plane anisotropy. The results are relevant for understanding the nuclear magnetic ordering in copper and silver at nanokelvin temperatures. Our work and the earlier spin-wave analysis are in accord with the observed type-I order in copper when the external magnetic field is aligned along the [001] and [110] crystalline directions, but in partial disagreement with the previous perturbation analyses. We investigate also an up-up-down spin configuration, which is consistent with the antiferromagnetic $(\frac{2}{3}\ \frac{2}{3}\ 0)$ Bragg reflection observed in copper. It has been proposed that the $(\frac{2}{3}\ \frac{2}{3}\ 0)$ order is stabilized by quantum fluctuations as theoretically calculated spin-spin interactions favor type-I modulation in the mean-field theory. We find, however, that quantum fluctuations favor type-I order rather than the $(\frac{2}{3}\ \frac{2}{3}\ 0)$ modulation. This result urges refined calculations of indirect nuclear-spin interactions in copper.

I. INTRODUCTION

The selection of the ground state in frustrated antiferromagnets has been investigated extensively in recent years. A classical problem in this field is the ground state of the fcc system.[1] Here we consider an assembly of spins interacting through an isotropic exchange coupling and an anisotropic dipolarlike force. We focus our attention on type-I antiferromagnetism,[2–14] characterized by ordering vectors $\mathbf{k}_1 = (\pi/a)(1,0,0)$, $\mathbf{k}_2 = (\pi/a)(0,1,0)$, and $\mathbf{k}_3 = (\pi/a)(0,0,1)$; $2a$ is the lattice constant. The ground-state spin order can be a single-**k** structure, which is described by only one of these **k** vectors, or a double-**k** or a triple-**k** state. For classical spins, these structures are degenerate. In an ideal system with no imperfections, a unique ground state at $T = 0$ is selected by quantum fluctuations.[3–7] We will study the ground state for various relative strengths of the isotropic and anisotropic interactions in zero field as well as in an external field.

We first investigate the effect of an external field on the ground state of the isotropic Heisenberg antiferromagnet. This model is particularly interesting because of its simplicity and the nontriviality of the results. It is perhaps surprising that this question has not been investigated before, at least not correctly, although a more complicated case, the one with easy-plane anisotropy, has been studied extensively.[4–6] We find for the isotropic model a transition from a single-**k** state to a triple-**k** structure with increasing field. The spin configurations in the triple-**k** region are rather similar to those obtained for two-dimensional triangular antiferromagnets; this reflects the frustration present in these lattices.

We then study the ground-state spin configurations in type-I fcc antiferromagnets with easy-plane anisotropy. The amplitudes of the antiferromagnetic modulations in these systems are perpendicular to the respective type-I ordering vectors. Such an anisotropy results from the dipolar interaction.[15] The calculations are relevant for an understanding of nuclear antiferromagnetic ordering in copper and silver, and possibly in gold and platinum as well.[16] Important tests for the theoretically calculated spin structures are imposed by recent neutron-diffraction measurements of copper below $T_N = 60$ nK.[17–19] At high fields in the antiferromagnetic region, experimental data allow one to discard some of the proposed spin structures. Although several theoretical calculations have been performed to determine the spin structures there have so far been serious discrepancies between the various approaches.[4–6,8–10] Our paper aims to clarify the situation.

We adopt the technique used by Lindgård[5,6,20] and Long[7] who both employed the second-order perturbation theory in order to investigate how quantum-mechanical spin fluctuations select the ground state among structures which are continuously degenerate for classical spins. We repeat and extend Lindgård's calculations[5,6,20] of the magnetic phase diagrams for easy-plane type-I fcc antiferromagnets. When the external field is oriented along the [110] crystalline directions, our results differ considerably from the previous calculations.[6] When $\mathbf{B} \parallel [001]$, our work differs from the earlier work[5] in high fields, which is the region relevant for a compari-

son against neutron-diffraction results. Our results are in agreement with the neutron-diffraction measurements unlike the previous perturbation calculations.[5,6] Our calculations are in good overall agreement with previous work[4] employing the linear spin-wave theory.

Finally, we study another problem associated with nuclear magnetic ordering in copper. The most recent neutron-diffraction measurements[18,19] have revealed in the intermediate fields of the antiferromagnetic region a $\mathbf{k}=(\pi/a)(2/3,2/3,0)$ modulation. It has been suggested by Lindgård[5,6,20] that this modulation is stabilized over the type-I order found in the high- and low-field regions by quantum fluctuations. Since our analysis of quantum effects for the type-I order differ significantly from his work,[5,6] it seems necessary to reinvestigate the problem of the phase at intermediate fields. Oppositely to the earlier results,[5,6,20] we find that quantum fluctuations favor type-I order rather than the $\mathbf{k}=(\pi/a)(2/3,2/3,0)$ modulation.

Our paper is organized as follows. In Sec. II, we specify the model and derive the equation for the quantum-fluctuation correction to the ground-state energy using perturbation theory. In Sec. III, we investigate the ground state of a type-I antiferromagnet with isotropic spin-spin interactions. In Sec. IV, we calculate the phase diagram for type-I antiferromagnets with easy-plane anisotropy when the external field is aligned along the [001] and the [110] crystalline axes. In Sec. V, we discuss the stability of the $\mathbf{k}=(\pi/a)(2/3,2/3,0)$ modulation vs type-I order. Section VI concludes our paper. In the Appendixes, we discuss the relationship between the present approach and the spin-wave theory, and compare our results in detail with earlier theoretical calculations.

II. THEORY

We assume for the Hamiltonian

$$H=\tfrac{1}{2}\sum_{i,j}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j +\tfrac{1}{2}\sum_{i,j}D_{ij}\{\mathbf{S}_i\cdot\mathbf{S}_j-3(\mathbf{S}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\mathbf{r}}_{ij}\cdot\mathbf{S}_j)\} -\mathbf{B}\cdot\sum_i\mathbf{S}_i\ , \tag{1}$$

which consists of exchange, dipolar, and the Zeeman terms, respectively. $\mathbf{S}_i$ is the spin operator acting at the lattice site i, J_{ij} and D_{ij} are the strengths of the exchange and dipolar forces, respectively, and $\mathbf{B}$ is the external magnetic field; $\hat{\mathbf{r}}_{ij}$ is a unit vector from spin i to spin j. The detailed form of the coupling constant $J_{ij}=J(|\mathbf{r}_i-\mathbf{r}_j|)$ is not important as long as it falls off with distance at least as fast as the dipolar interaction. We assume further that the spin-spin interactions are such that the type-I antiferromagnetic order is favored.

We rewrite the Hamiltonian of Eq. (1) more compactly as

$$H=\tfrac{1}{2}\sum_{i,j}\mathbf{S}_i\underline{A}_{ij}\mathbf{S}_j-\mathbf{B}\cdot\sum_i\mathbf{S}_i\ , \tag{2}$$

where the interaction matrix $A_{ij}^{\mu\nu}=J'_{ij}(\delta^{\mu\nu}-d_{ij}\,\hat{\mathbf{r}}_{ij}^{\mu}\hat{\mathbf{r}}_{ij}^{\nu})$ has been introduced. Parameters J'_{ij} and d_{ij} are defined by $J'_{ij}=J_{ij}+D_{ij}$ and $d_{ij}=3D_{ij}/J'_{ij}$. When i and j are nearest neighbors, d_{ij} is denoted by $d=3D_1/(J_1+D_1)$. We adopt the quantity d as a measure of the relative strength of the dipolar interaction.

The Hamiltonian of Eq. (2) is usually analyzed by the use of the Fourier transformation of the interaction matrix $\underline{A}(\mathbf{k})=\sum_j\underline{A}_{ij}e^{-i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{r}_j)}$.[21,22] The lowest energy for ordering is found by inspecting the eigenvalues $\lambda_{\mathbf{k},n}$ and the eigenvectors $\mathbf{x}_{\mathbf{k},n}$ of $\underline{A}(\mathbf{k})$.

A. Type-I order

A type-I spin structure is characterized by the ordering vectors $\mathbf{k}_1=(\pi/a)(1,0,0)$, $\mathbf{k}_2=(\pi/a)(0,1,0)$, and $\mathbf{k}_3=(\pi/a)(0,0,1)$, which are parallel to the crystalline axes of the fcc lattice. Let us denote the lowest eigenvalue of the 3×3 matrix $\underline{A}(\mathbf{k})$ by $\lambda_{\mathbf{k}}$ and the smallest $\lambda_{\mathbf{k}}$ by λ. We assume that λ is equal to $\lambda_{\mathbf{k}}$ only when $\mathbf{k}$ is one of the three type-I ordering vectors $\mathbf{k}_j$, $j=1,2,3$. It then follows that the ground state for classical spins can be written as

$$\mathbf{S}_i^{\mathrm{cl}}/S=\mathbf{m}+\sum_{j=1,2,3}\mathbf{d}_j\cos(\mathbf{k}_j\cdot\mathbf{r}_i)\ , \tag{3}$$

where the vector $\mathbf{d}_j$ is the amplitude of the antiferromagnetic modulation with a wave vector $\mathbf{k}_j$. The magnetization $\mathbf{m}=\mathbf{B}/B_c$ is proportional to the external field B, with $B_c=S[\lambda(\mathbf{k}=\mathbf{0})-\lambda]$ the critical field for type-I antiferromagnetism at $T=0$. It follows from Eq. (3) that there are, in general, four sublattices in a type-I fcc antiferromagnet.

The anisotropy of the dipolar interaction imposes the constraints[21]

$$\mathbf{d}_j\cdot\mathbf{k}_j=0\ ,\quad j=1,2,3\ , \tag{4}$$

for the ground-state spin configuration. Thus the anisotropy has the easy-plane character. The equation follows from the nature of the eigenvectors $\mathbf{x}_{\mathbf{k}_j,n}$ of the matrix $\underline{A}(\mathbf{k}_j)$: It is equivalent to the requirement that the vector $\mathbf{d}_j$ belongs to the eigenspace of the eigenvalue λ.

Requiring $|\mathbf{S}_i^{\mathrm{cl}}|=S$ for all spins i, one finds the conditions[22]

$$|\mathbf{m}|^2+|\mathbf{d}_1|^2+|\mathbf{d}_2|^2+|\mathbf{d}_3|^2=1\ , \tag{5a}$$

$$\begin{aligned}&\mathbf{m}\cdot\mathbf{d}_1+\mathbf{d}_2\cdot\mathbf{d}_3=0,\\ &\mathbf{m}\cdot\mathbf{d}_2+\mathbf{d}_3\cdot\mathbf{d}_1=0\ ,\\ &\mathbf{m}\cdot\mathbf{d}_3+\mathbf{d}_1\cdot\mathbf{d}_2=0\ .\end{aligned} \tag{5b}$$

Equation (4) and (5) define, for classical spins, a two-parameter set of continuously degenerate states with the energy

$$E_0^{\rm cl} = \tfrac{1}{2}NS^2\{\lambda - [\lambda(\mathbf{k}=0) - \lambda]m^2\} \ . \tag{6}$$

The degeneracy is lifted, however, by thermal and/or quantum fluctuations.[3–12]

B. Perturbative calculation of quantum fluctuations

For notational convenience, we replace the spin operators with bosonic creation and annihilation operators $a_i^\dagger$, a_i using the Holstein-Primakoff transformation[23]

$$\begin{aligned}\mathbf{S}_i = {} & \mathbf{e}_z^i(S - a_i^\dagger a_i) + \mathbf{e}_+^i a_i^\dagger (S - a_i^\dagger a_i/2)^{\frac{1}{2}} \\ & + \mathbf{e}_-^i (S - a_i^\dagger a_i/2)^{\frac{1}{2}} a_i \ ,\end{aligned} \tag{7}$$

where $\mathbf{e}_z^i = \underline{R}_i \mathbf{e}_z$, $\mathbf{e}_\pm^i = \underline{R}_i \mathbf{e}_\pm$, and $\mathbf{e}_\pm = (1/\sqrt{2})(\mathbf{e}_x \pm i\mathbf{e}_y)$; $\underline{R}_i$ is a rotation matrix defined so that the unit vector $\mathbf{e}_z^i$ coincides with the classical spin direction $\mathbf{S}_i^{\rm cl}$. Now we can write the Hamiltonian as $H = H_0 + \Delta H$, where operators H_0 and ΔH have the expressions

$$H_0 = E_0^{\rm cl} + \sum_j \mathbf{B}_j^{\rm loc} \cdot \mathbf{e}_z^j \ a_j^\dagger a_j, \tag{8a}$$

$$\begin{aligned}\Delta H = {} & -\sqrt{S}\sum_j \mathbf{B}_j^{\rm loc} \cdot \mathbf{e}_+^j \ a_j^\dagger \\ & + \tfrac{1}{2}S\sum_{i,j}\{\mathbf{e}_+^i \underline{A}_{ij} \mathbf{e}_+^j \ a_i^\dagger a_j^\dagger + \mathbf{e}_+^i \underline{A}_{ij} \mathbf{e}_-^j \ a_i^\dagger a_j\} + \text{H.c.} \\ & + \text{higher-order terms.}\end{aligned} \tag{8b}$$

The local field is[22]

$$\mathbf{B}_j^{\rm loc} \equiv \mathbf{B} - S\sum_i \mathbf{e}_z^i \underline{A}_{ij} = -\lambda S \mathbf{e}_z^j \ , \quad B \le B_c \ . \tag{9}$$

Corrections to the noninteracting spin-wave approximation are included in the higher-order terms.

To estimate the quantum correction for the ground-state energy of H_0 we note first that the linear terms in the bosonic operators vanish because $\mathbf{e}_z^j \cdot \mathbf{e}_\pm^j = 0$. Applying the second-order perturbation theory we find

$$\begin{aligned}\delta E_2 = {} & \frac{1}{E_0 - E_2}\tfrac{1}{2}\sum_{i\neq j} |\langle 0|a_j a_i \Delta H|0\rangle|^2 \\ = {} & -\frac{S^2}{E_2 - E_0}\tfrac{1}{2}\sum_{i,j} |\mathbf{e}_+^i \underline{A}_{ij} \mathbf{e}_+^j|^2 \ .\end{aligned} \tag{10}$$

The difference between the ground-state energy E_0 and the excited-state energy E_2 corresponds to the creation of a spin deviation at two different sites, i and j, yielding $E_2 - E_0 = -2\lambda S$. Note that the higher terms of Eq. (8b) do not enter in this order of perturbation theory.

One expects that the terms in the sums of Eq. (10) fall off like r_{ij}^{-6}. Owing to this rapid decay the sums are approximated rather well by the nearest-neighbor terms only as has been pointed out by Lindgård.[5,6,20] One can estimate that the contribution of the nearest-neighbor terms is over 80% of the total for a fcc lattice. Moreover, when the anisotropic part of the interactions vanishes the relation

$$\sum_{\text{all pairs}} = C(\{J'_{ij}\}) \sum_{\text{NN only}} \tag{11}$$

holds exactly; C depends only on the coupling constants and the lattice geometry. Therefore one can quite safely ignore the contribution of the more distant couplings to δE_2. Our major approximation is thus the truncation of the perturbative series at the second order.

The leading correction to the classical energy can be written as

$$\delta E_2 = c \ \Sigma_2 \ , \tag{12}$$

where $c = -NSJ_1'^2/(2\lambda)$ and Σ_2 is given by the dimensionless sum

$$\Sigma_2 = -\frac{1}{NJ_1'^2}\tfrac{1}{2}\sum_{\langle i,j\rangle} |\mathbf{e}_+^i \underline{A}_{ij} \mathbf{e}_+^j|^2 \ , \tag{13}$$

which involves the nearest-neighbor pairs only. Among the degenerate states, the configuration that minimizes Σ_2 is the ground state.

III. TYPE-I ORDER FOR ISOTROPIC SPIN-SPIN INTERACTIONS

We first investigate the ground state of the type-I fcc antiferromagnet with isotropic spin-spin interactions, i.e., the Heisenberg model. We write Eq. (13) in terms of the spin vectors by choosing $\mathbf{e}_+^i = (1/\sqrt{2})(\mathbf{u} + i\mathbf{v}_i)$ and $\mathbf{e}_+^j = (1/\sqrt{2})(\mathbf{u} + i\mathbf{v}_j)$, where $\mathbf{u} = S^{-2}\mathbf{S}_i \times \mathbf{S}_j/[1 - S^{-4}(\mathbf{S}_i \cdot \mathbf{S}_j)^2]^{1/2}$, $\mathbf{v}_i = S^{-1}\mathbf{S}_i \times \mathbf{u}$, and $\mathbf{v}_j = S^{-1}\mathbf{S}_j \times \mathbf{u}$. Noting that $\mathbf{v}_i \cdot \mathbf{v}_j = S^{-2}\mathbf{S}_i \cdot \mathbf{S}_j$, we obtain

$$\begin{aligned}\Sigma_2 = {} & -\frac{1}{4N}\tfrac{1}{2}\sum_{\langle i,j\rangle}[1 - S^{-2}(\mathbf{S}_i \cdot \mathbf{S}_j)]^2 \\ = {} & -\frac{5}{2} + 4m^2 - \frac{1}{4S^4}\sum_{a>b}(\mathbf{S}_a \cdot \mathbf{S}_b)^2 \ ,\end{aligned} \tag{14}$$

where the sum in the last equation is taken over the four sublattices a, b. The result is in agreement with Larson and Henley[24] and, except for a factor of 2 and a constant, with Long.[7] The equation shows that collinear structures are favored.[11,25] In zero field the stable structure is the single-**k** state with sublattice spin directions $\mathbf{S}_1 = \mathbf{S}_2 = -\mathbf{S}_3 = -\mathbf{S}_4$. Similarly, at $B = B_c/2$ the collinear triple-**k** structure of Fig. 1(c), with $\mathbf{S}_1 = \mathbf{S}_2 = \mathbf{S}_3 = -\mathbf{S}_4$ and $\mathbf{S}_a \parallel \mathbf{B}$, is the ground state.

We find that the phase diagram consists of a single-**k** structure at low fields and a triple-**k** state at high fields. The transition occurs at $m = B/B_c = 0.407$. The spins in the four sublattices are always coplanar; the evolution of the spin configurations with increasing field is illustrated in Fig. 1. In the single-**k** state, Fig. 1(a), the spins simply tilt towards the field. In the triple-**k** structure at $0.407 < m < 0.5$, Fig. 1(b), spins in three sublattices are tilted towards the field so that two of these sublattices have the same spin direction; the spins in the fourth sublattice are nearly opposite to the magnetic field. In the

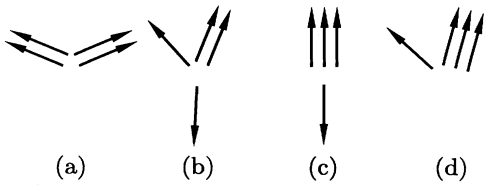

FIG. 1. Directions of the spins in the four sublattices of type-I structure in an external field (upwards); spin-spin interactions are isotropic. The single-**k** (a) and the triple-**k** (b) structures at the transition point $m \approx 0.407$. The triple-**k** state when $m = 0.5$ (c) and when $m = 0.9$ (d).

triple-**k** structure at $m \geq 1/2$, Figs. 1(c) and 1(d), the spins in three sublattices have same directions, while the fourth sublattice adjusts itself to match $\mathbf{m} = \mathbf{B}/B_c$.

A planar triangular (pt) lattice can be regarded as a two-dimensional counterpart of a fcc structure. The classical ground state of the Heisenberg[26] and the XY[27,28] models with nearest-neighbor interactions in the pt lattice is a three-sublattice structure with an infinite degeneracy, but a unique ground state is selected by quantum fluctuations.[29] Applying our present results to these models, the ground state is found to minimize the function $-[(\mathbf{S}_1 \cdot \mathbf{S}_2)^2 + (\mathbf{S}_2 \cdot \mathbf{S}_3)^2 + (\mathbf{S}_3 \cdot \mathbf{S}_1)^2]$, where $\mathbf{S}_a$ is the spin direction in sublattice a. The minimization, which has been carried out in Ref. 30, yields three different coplanar spin structures with increasing field. These configurations are similar to our results for isotropic type-I fcc antiferromagnets. The spin patterns for the pt lattice are obtained from Figs. 1(b)–1(d) if one removes one of the parallel vectors from each diagram, and adjusts slightly the spin vectors in the diagrams (b) and (d). In particular, the collinear up-up-up-down structure (c) of Fig. 1 has an up-up-down counterpart in the pt lattice. The up-up-down configuration has been found to be stable in a large region of the (T, B) phase diagram in intermediate magnetic fields.[26–28] The ordering temperature is in fact highest in a finite field and corresponds to the up-up-down configuration. Similarly, we expect that the collinear up-up-up-down structure occupies an appreciable region in the (T, B) diagram of the isotropic type-I fcc antiferromagnet.

IV. TYPE-I ORDER WITH EASY-PLANE ANISOTROPY

For anisotropic spin-spin interactions the spin vectors are coupled to the crystalline directions. The various spin structures can be illustrated by drawing the projections of the spin vectors on a plane perpendicular to the magnetic field, as shown in Figs. 2 and 3.

We apply Eq. (13) given by the real-space perturbation theory in order to calculate the ground state of an easy-plane type-I fcc antiferromagnet in a magnetic field. There are 12 different terms in the sum of Eq. (13). All nearest neighbors of a spin site belonging to the first sublattice are shown in Fig. 2. Bonds between spin 1 and its nearest neighbors give six of the terms contributing

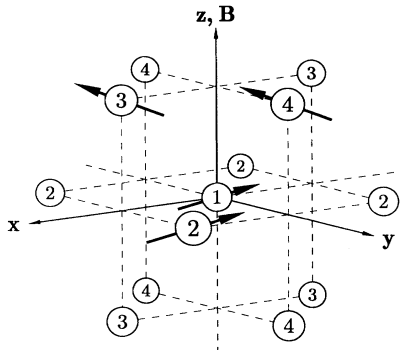

FIG. 2. Nearest neighbors of a spin on site 1 in a fcc lattice. The structure is the **1a** phase of Fig. 3.

to Σ_2; the remaining six involve the other sublattices. To evaluate Eq. (13) we first solve Eqs. (4) and (5). We then construct the spin vectors for the four sublattices and calculate the $\mathbf{e}_+^i$ vectors and finally write down the different terms contributing to Σ_2. We search through all possible type-I structures; this was not done in the earlier works.[4–6,20]

A. Phase diagram in fields along [001]

Description of our calculation is easier if we first present the resulting phase diagram, shown in Fig. 3. It contains four different phases. They are labeled according to the number of nonzero vectors $\mathbf{d}_j$. The single-**k** and double-**k** phases **1a**, **1b**, and **2** are described by spin projection diagrams of the same form at all fields. This is due to the orthogonality of vectors $\mathbf{d}_j$ and $\mathbf{m}$; the effect of the magnetic field is only to alter the length of every spin projection in the same way. For a triple-**k** struc-

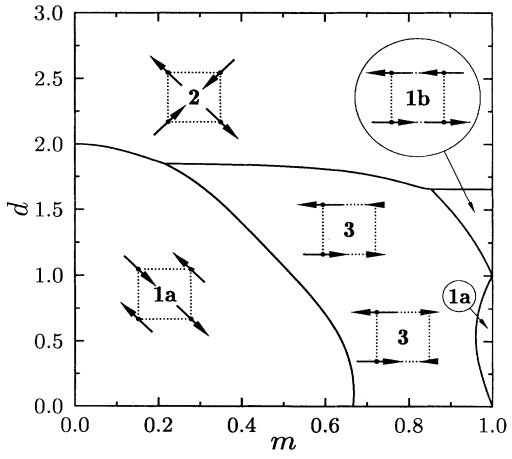

FIG. 3. Phase diagram of easy-plane type-I fcc antiferromagnets in a magnetic field oriented along a [001] crystalline direction. The coordinates in the figure are the magnetization $m = B/B_c$ and the parameter $d = 3D_1/(J_1 + D_1)$ expressing the strength of the nearest-neighbor dipolar interaction D_1 over the exchange force J_1. The various spin configurations have been illustrated by projection diagrams with labels indicating the number of **k** vectors in the structure.

ture, vectors $\mathbf{d}_j$ and $\mathbf{m}$ are necessarily nonperpendicular and the projection diagram is generally a function of the magnetic field. The descriptions of the triple-**k** states **3** in Fig. 3 are very schematic because of this fact.

The expressions for the spin configuration appearing in the diagram are

$$\mathbf{1a}: \mathbf{d}_3 = \frac{p}{\sqrt{2}}(\mathbf{e}_x + \mathbf{e}_y), \tag{15a}$$

$$\mathbf{1b}: \mathbf{d}_1 = p\,\mathbf{e}_y; \tag{15b}$$

$$\mathbf{2}: \begin{cases} \mathbf{d}_1 = (p/\sqrt{2})\,\mathbf{e}_y, \\ \mathbf{d}_2 = (p/\sqrt{2})\,\mathbf{e}_x; \end{cases} \tag{15c}$$

$$\mathbf{3}: \begin{cases} \mathbf{d}_1 = p\,\sin\theta\cos\phi\,\mathbf{e}_y, \\ \mathbf{d}_2 = p\,\sin\theta\sin\phi\,\mathbf{e}_z, \quad \tan\phi = -\dfrac{p}{m}\cos\theta, \\ \mathbf{d}_3 = p\,\cos\theta\,\mathbf{e}_y \end{cases} \tag{15d}$$

where $p = \sqrt{1-m^2}$.

We cannot, in general, express the triple-**k** state **3** compactly as a function of d and m. For $d = 0$, $d = 1$ and $m = 1/2$ it is, however, possible to express the lowest energy structure, if it belongs to the set Eq. (15d), as

$$\theta = \arccos\sqrt{\frac{m}{m+1}} \tag{16}$$

(drawn also in Fig. 4). Equation (15d) yields then

$$\mathbf{d}_1 = \mathbf{d}_3 = [m(1-m)]^{1/2}\mathbf{e}_y, \qquad \mathbf{d}_2 = (m-1)\mathbf{e}_z, \tag{17}$$

and the spins in the four sublattices are [Eq. (3)]

$$\begin{aligned} \mathbf{S}_1/S &= 2[m(1-m)]^{1/2}\mathbf{e}_y + (2m-1)\mathbf{e}_z, \\ \mathbf{S}_2 &= S\mathbf{e}_z, \\ \mathbf{S}_3/S &= -2[m(1-m)]^{1/2}\mathbf{e}_y + (2m-1)\mathbf{e}_z, \\ \mathbf{S}_4 &= \mathbf{S}_2, \end{aligned} \tag{18}$$

where the subscripts 1–4 refer to the sublattices indicated in Fig. 2. Spins in two of the sublattices are parallel to the field whereas spins in the other two tilt towards the field when **B** is increased. Following Lindgård[5] we use the notation $\leftrightarrows\,\circ\,\circ$ for this symmetric triple-**k** state. We find in agreement with him that the triple-**k** structure is stable for $m > 2/3$ when $d = 0$. The phase **3** acquires, however, the symmetric form $\leftrightarrows\,\circ\,\circ$ only at $d = 0$, $d = 1$, and $m = 1/2$ in disagreement with Ref. 5.

Figure 4 shows, for four values d, the parameter θ corresponding to the structure **3** as a function of m; the curves intersect at $m = 1/2$. For $d = 1.5$ there is a continuous transition **3** → **1b** at high fields. When $d < 1$ the high-field transition **3** → **1a** is of first order. It is obvious that the phase **3** in general differs considerably from the symmetric form $\leftrightarrows\,\circ\,\circ$ illustrated by the curve for $d = 1$.

The results of Fig. 3 are in a fairly good agreement with the previous spin-wave calculation[4] except for the structure **3** which was not investigated in Ref. 4 (see Appendix B for a detailed discussion).

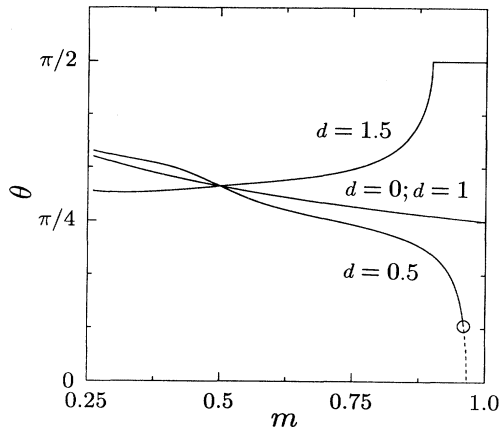

FIG. 4. Angle θ of the lowest energy triple-**k** structure, Eq. (15d), for four values of the parameter d as a function of the magnetization $m = B/B_c$. The topmost ($d = 1.5$) curve shows a continuous transition (at the kink) to the **1b** state; the point where the **1a** state becomes more favorable is marked on the lowest ($d = 0.5$) curve. See Fig. 3 for transition points in low fields.

When comparing our results with the previous perturbation calculation of Lindgård,[5] we find an exact agreement along the lines $d = 0$ and $m = 0$ and a good overall agreement for $m < 0.8$. In high fields, $m > 0.8$, there are considerable differences (see Appendix B). These differences are crucial in a comparison against experiments on copper.

In recent neutron-diffraction measurements[17–19] of nuclear ordering in copper, the antiferromagnetic (1 0 0) Bragg reflection was measured while the external field was decreased to zero starting from a field larger than the critical field for antiferromagnetic order, $B_c = 0.25$ mT. In the field region $B = 0.16 - 0.25$ mT, the observed (1 0 0) intensity showed a strong dependence on the alignment of the external field (Figs. 13 and 17 in Ref. 19): The (1 0 0) intensity was strong when the field was perpendicular to the respective ordering vector, i.e., when $\mathbf{B} \parallel [001]$, but almost zero when the field was parallel to this vector, $\mathbf{B} \parallel [100]$. Both experiments[31] and calculations[32,33] yield $d \approx 2$ for nearest-neighbor (NN) copper nuclei. Therefore the stable structure in high fields is either **2** or **1b** according to Fig. 3. When a comparison is made with the experiment, it is necessary to consider an average over all forms of the stable structure allowed by the symmetry: These are realized as different domains in the specimen. For the structure **2** [Eq. (15c)], the perpendicular and parallel reflections are proportional to the domain averages $\frac{1}{2}(|\mathbf{d}_1|^2 + |\mathbf{d}_2|^2) = \frac{1}{2}(1-m^2)$ and $|\mathbf{d}_3|^2 = 0$, respectively. Thus the neutron intensities are in qualitative agreement with the state **2**, predicted by the spin-wave calculations.[4] The high-field structure of the previous perturbation calculations[5] is in disagreement with the neutron-diffraction results.

Although the structure **1b** would also yield the observed neutron-diffraction results immediately below $B = B_c$, discrepancies would occur at lower fields close

to 0.16 mT where the triple-**k** structure **3** becomes stable according to Fig. 3. The parallel reflection should then clearly deviate from zero in disagreement with the neutron-diffraction data.[19] In fields below 0.16 mT, neutron-diffraction measurements show the presence of the $(\pi/a)(2/3,2/3,0)$ ordering vector in addition to the type-I vector and therefore a meaningful comparison with the present results can be made only at high fields. In zero field, the three different type-I modulations should be equivalent making it impossible to distinguish between different type-I structures.

B. [110] field direction

Equations (4) and (5) were solved again for all possible type-I spin structures with easy-plane anisotropy. There are two branches of solutions. The first branch is of the general form

$$\textbf{3b:}\quad \begin{cases} \mathbf{m} &= m(\mathbf{e}_x+\mathbf{e}_y)/\sqrt{2}, \\ \mathbf{d}_1 &= d_{1z}\mathbf{e}_z, \\ \mathbf{d}_2 &= d_{2z}\mathbf{e}_z, \\ \mathbf{d}_3 &= d_{3x}\mathbf{e}_x + d_{3y}\mathbf{e}_y, \end{cases} \tag{19}$$

and the second branch can be solved assuming $d_{2x} \neq 0$. The solutions for the triple-**k** structures can be expressed as functions of, say, d_{2z} and d_{3x}. Our numerical studies show that all stable triple-**k** structures reside in the branch of Eq. (19). The resulting phase diagram is shown in Fig. 5.

The largest area of the figure is occupied by the structure

$$\textbf{3a:}\quad \begin{cases} d_{1z} = d_{2z} = [m(1-m)]^{1/2}, \\ d_{3x} = d_{3y} = (m-1)/\sqrt{2}, \end{cases} \tag{20}$$

found previously in Ref. 4. The configuration **3a** is a special case of the more general structure **3b**. Using Eq. (3) one obtains for the spin vectors of **3a**

$$\textbf{3a:}\quad \begin{cases} \mathbf{S}_1/S &= (2m-1)(\mathbf{e}_x+\mathbf{e}_y)/\sqrt{2} \\ &\quad +2[m(1-m)]^{1/2}\mathbf{e}_z, \\ \mathbf{S}_2/S &= (2m-1)(\mathbf{e}_x+\mathbf{e}_y)/\sqrt{2} \\ &\quad -2[m(1-m)]^{1/2}\mathbf{e}_z, \\ \mathbf{S}_3/S &= (1/\sqrt{2})(\mathbf{e}_x+\mathbf{e}_y), \\ \mathbf{S}_4 &= \mathbf{S}_3, \end{cases} \tag{21}$$

where the sublattices 1–4 are defined in Fig. 2. This structure resembles the symmetric form Eq. (18) of the configuration **3** for $\mathbf{B} \parallel [001]$: It is a four-sublattice triple-**k** state for which spins in two sublattices are parallel to the field for all $B < B_c$. Configuration **3a** is always of the symmetric form and therefore different in its nature from structure **3** for $\mathbf{B} \parallel [001]$. The states **3a** and **3** are exactly identical at the isotropic limit $d = 0$ because the coupling between the spin directions and the crystalline axes then vanishes.

At zero field the spin configuration **2** is stable when $d > 2$. This corresponds to (e.g.) $d_{2z} = d_{3y}$, $d_{1z} = d_{3x} = 0$ in Eq. (19). The configuration **3b** of Fig. 5 can

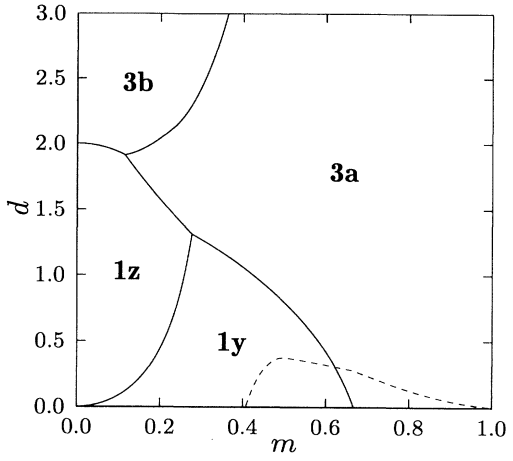

FIG. 5. Phase diagram for the magnetic field in the [110] direction as a function of $m = B/B_c$ and $d = 3D_1/(J_1 + D_1)$. Below the dashed line quantum effects tend to stabilize the triple-**k** structure of Figs. 1(b)–1(d) in a $S = \frac{1}{2}$ system.

be considered as an extension of **2** at fields along [110]: When $B \to 0$ d_{1z} and d_{3x} vanish continuously. When $B \neq 0$ the coefficients $d_{j\alpha}$ in Eq. (19) for **3b** are nonzero and different from each other. At a low intermediate field a discontinuous transition takes place to the symmetric configuration **3a**.

Finally, in the lower left corner of Fig. 5 the single-**k** structures **1z** and **1y** are stable. For these the nonzero vectors $\mathbf{d}_j$ are given by

$$\textbf{1z:}\quad \mathbf{d}_3 = (p/\sqrt{2})(\mathbf{e}_x - \mathbf{e}_y), \tag{22a}$$

$$\textbf{1y:}\quad \mathbf{d}_2 = p\mathbf{e}_z\ , \tag{22b}$$

where $p = \sqrt{1-m^2}$. We find that the **1z** phase has lower energy than the **1y** phase when $m < \sqrt{3/29} \approx 0.32$ and $d > 8m^2/(1-7m^2)$. In zero field **1z** is identical to **1a** [Eq. (15a)] and **1y** is the same as **1b**.

The results of Fig. 5 are in fairly good agreement with the spin-wave calculations by Viertiö and Oja.[4] When comparing our result with the previous perturbation calculation of Lindgård,[6,20] we find considerable differences. In fact most parts of our diagram differ from his result. (See Appendix B for details.)

In copper $d \approx 2$, and the high-field structure should be the triple-**k** structure **3a**. Equation (20) predicts that both the (1,0,0) and (0,1,0) neutron-diffraction intensities should be proportional to the domain average $\frac{1}{2}(|\mathbf{d}_1|^2 + |\mathbf{d}_2|^2) = m(1-m)$. The domain average for the (0,0,1) intensity should be proportional to $|\mathbf{d}_3|^2 = (m-1)^2$ and therefore it should be much weaker than the (1,0,0) and (0,1,0) reflections at high fields. The neutron-diffraction data[19] (Figs. 13 and 17 in Ref. 19) is in agreement with the behavior for the **3a** state, found previously in the spin-wave calculation.[4] To compare theory against experiments in lower fields, one has to consider interplay between the (1,0,0) and $(\frac{2}{3},\frac{2}{3},0)$ modulations.[34–36]

C. $d \to 0$ limit

In the $d \to 0$ limit the results for the anisotropic model should be identical to those found for the isotropic case. This is, however, not the case. The reason is that the limit $d \to 0$ is artificial in the sense that we still require the conditions of Eqs. (4) due to the anisotropy of the interactions. This way we do not explore all possible triple-**k** structures. For single-**k** and double-**k** states this yields no loss of generality because **m** and the nonzero vectors $\mathbf{d}_j$ constitute an orthogonal basis, and only the degree of freedom associated with the rotation about the field is ignored. It seems that the previous calculations[4–6] for the easy-plane type-I fcc antiferromagnets fail to yield the correct spin structures at $d \to 0$ for these reasons. Moreover, in these calculations the spin structures depended at the isotropic limit on the direction of the field with respect to the crystalline axes: This is clearly an unphysical feature.

In order to find the ground state at small d we note first that the triple-**k** phase violates the constraints $\mathbf{d}_j \cdot \mathbf{k}_j = 0$ of an easy-plane system [Eqs. (4)] and cannot be stable in the presence of an appreciable dipolar anisotropy. For example, when $m = 1/2$ the modulation vectors are $\mathbf{d}_1 = \mathbf{d}_2 = \mathbf{d}_3 = -\mathbf{m} = -\mathbf{B}/B_c$, and Eqs. (4) remain unsatisfied by any choice of the field direction.

When the anisotropy is weak the ground state obtained for the fully isotropic interaction is, however, approximately correct and features of the isotropic case extend into the (m, d) phase diagram of the anisotropic model. An accurate treatment of the $d \to 0$ limit is difficult because at small d quantum fluctuations stabilize a structure which does not minimize the classical energy. This leads to appearance of negative or complex excitation energies in a conventional spin-wave calculation.[37,38] Analogously to the calculations by Rastelli, Reatto, and Tassi[37] for the ferrohelix transition line in the Heisenberg model, we estimate the crossover between the ground states obtained for isotropic and anisotropic models by examining when the increase in the anisotropy energy for the triple-**k** structures of Figs. 1(b)–1(d) due to a finite d equals the energy difference the triple-**k** structure gains in Σ_2 (evaluated at $d = 0$). This defines a phase line in the (m, d) plane which depends on the spin S because we are comparing classical and quantum effects.

At $0.407 < m < 0.667$ we compare the triple-**k** structure with a single-**k** structure [$\Sigma_2 = -4(1 - m^2)^2$]. At $0.667 < m < 1$ the triple-**k** structure is compared with the $\leftrightarrows$ state for which $\Sigma_2 = -4(1 - m)^2(1 + 4m^2)$ at $d = 0$. Σ_2 for the triple-**k** state is

$$m \geq \tfrac{1}{2}\text{: } \Sigma_2 = -16(1 - m^2)^2/3 \ , \tag{23a}$$

$$m \leq \tfrac{1}{2}\text{: } \Sigma_2 = -4(1 + m)^2 \times (1 + 4m - 16m^3 + 32m^4)/(1 + 4m)^2 \ . \tag{23b}$$

The latter equation is an approximation where we have assumed that the spin in Fig. 1(b) which is nearly opposite to the field is *exactly* opposite to **B**. In the stability region the error is less than 2×10^{-3}.

The result of this estimation for $S = \frac{1}{2}$ is shown in Fig. 5. In the region below the dashed line quantum fluctuations stabilize the triple-**k** state of Figs. 1(b)–1(d). The region collapses towards the line $d = 0$ in the (m, d) diagram with increasing S. For a given m, the quantity d at the phase boundary of this region is proportional to S^{-1}. The boundary of the quantum-fluctuation-stabilized phase depends only weakly on the field direction. It has been left out from Fig. 3 for clarity.

D. Application to materials other than Cu

In silver, three different NMR measurements[39–41] and a band-structure calculation[42] yield consistently $d = 0.75 \pm 0.06$. Thus we predict three phases for both $\mathbf{B} \parallel [001]$ and $\mathbf{B} \parallel [110]$. The most pronounced features of the phase diagrams are the single-**k** $\to$ triple-**k** transitions in intermediate fields. NMR measurements of a polycrystalline silver sample can be interpreted in terms of two ordered structures as a function of the field.[43] A detailed comparison of theory and experiments must wait, however, for neutron-diffraction experiments.

The predicted phase diagram for nuclear spins in gold, $S = \frac{3}{2}$, is similar to that of silver if the exchange interactions scale simply according to the measured hyperfine splitting, yielding $d = 0.07$.[4] In this case the anisotropy would be so weak that the triple-**k** states would resemble those of the isotropic model. The uncertainty in anisotropic exchange interactions is, however, large preventing reliable predictions. Uncertainties in exchange interactions are large also in platinum and rhodium,[44] which are fcc nuclear magnets of current experimental interest.

Neither can we apply our results to the electronic fcc type-I antiferromagnets $MnTe_2$ (Ref. 13) and UO_2.[14] Although the isotropic exchange interactions in $MnTe_2$ have been calculated theoretically[45] and also inferred from measurements,[46] the anisotropic interaction has not yet been determined. In UO_2, on the other hand, the spin-lattice interaction, which is not included in our model, is important.[14]

V. TYPE-I vs $\mathbf{k} = (\pi/a)(2/3, 2/3, 0)$ ORDER IN COPPER

Neutron-diffraction experiments on copper have revealed an unusual antiferromagnetic structure, characterized by the Bragg reflection $(\pi/a)(2/3, 2/3, 0)$ in intermediate fields $B = 0.01 - 0.13$ mT below the critical field $B_c = 0.25$ mT.[18] As this kind of order has not been observed in a fcc system before, the origin of the phase and its spin structure have been investigated in several theoretical calculations.[5,6,20,34–36] It has been suggested by Lindgård[5,6,20] that quantum fluctuations favor the $\mathbf{k} = (\pi/a)(2/3, 2/3, 0)$ modulation in intermediate fields rather than the type-I order found in the high- and low-field regions. According to his original argument,[5,6] quantum fluctuations should make $\mathbf{k} = (\pi/a)(\eta, \eta, 0)$ phase penetrate between the **1z** and **1y** states in interme-

diate fields. Our diagram of Fig. 5 gives no justification for this particular mechanism as the **1z** $\leftrightarrow$ **1y** boundary does not extend up to the d values relevant for copper, $d \approx 2$, as it did in Lindgård's phase diagram.[6] It is therefore necessary to reinvestigate the subject.

We assume for simplicity that the $(\pi/a)(1,0,0)$ and $(\pi/a)(2/3,2/3,0)$ orders are degenerate; i.e., their eigenvalues are equal, so that the values of Σ_2 are directly comparable. We study the case when the external field is along the [110] direction and $B = B_c/3$. The observed $(\pi/a)(2/3,-2/3,0)$ Bragg reflection is strongest around this field.[18] Among the spin configurations modulated by the 12 vectors in the star of $(\pi/a)(2/3,-2/3,0)$, the lowest energy corresponds to the structure[34,47]

$$\mathbf{S}_i^{\rm cl}/S = \frac{\sqrt{2}}{6}(\mathbf{e}_x + \mathbf{e}_y)\left\{1 - 4\cos\left[\frac{2\pi}{3a}(\mathbf{e}_x - \mathbf{e}_y)\cdot\mathbf{r}_i\right]\right\}, \tag{24}$$

which satisfies $|\mathbf{S}_i| = S$ and involves only $(\pi/a)(2/3,-2/3,0)$ order and a constant magnetization. The spin configuration has the up-up-down pattern: $\uparrow\uparrow\downarrow$. We find for this state

$$\Sigma_2 = -(160 - 128d + 39d^2)/48 . \tag{25}$$

The analogous result for the type-I structure **3a** of Eq. (20) is

$$\Sigma_2 = -(3328 - 2528d + 965d^2)/1296 , \tag{26a}$$

and for the single-**k** structure **1y** of Eq. (22b)

$$\Sigma_2 = -(4096 - 3136d + 941d^2)/1296 . \tag{26b}$$

Adopting $d = 2$ for copper we find that $\Sigma_2 = -1.25$ for $\uparrow\uparrow\downarrow$, -1.65 for **3a**, and -1.23 for **1y**. Thus the type-I structure **3a** is favored by quantum fluctuations.

One should note that although collinear structures such as the $\uparrow\uparrow\downarrow$ configuration are favored by quantum fluctuations when the interactions are isotropic,[25] this is not the case for sufficiently anisotropic interactions like those in copper.

Our present calculation gives support to the idea advocated by Viertiö and Oja[34] who explained the observed stability of the $\mathbf{k} = (\pi/a)(2/3,2/3,0)$ order by starting from the assumption that the exchange interactions are such that the eigenvalue for $\mathbf{k} = (\pi/a)(2/3,2/3,0)$ is in fact lower than that for a type-I ordering vector. The question to address is why type-I order, rather than the $\mathbf{k} = (\pi/a)(2/3,2/3,0)$ modulation, is observed in low and high fields; a solution has been constructed by using the mean-field theory.[34–36] The problem remaining is that the eigenvalue for the $\mathbf{k} = (\pi/a)(2/3,2/3,0)$ order is 10% higher than that for the type-I order according to first-principles band-structure calculations.[32,33] As the 10% difference in the two eigenvalues is within the estimated uncertainty of the calculated exchange parameters, there is no serious controversy. Nevertheless, refined calculations of the exchange interactions in copper would be worthwhile.

VI. CONCLUSIONS

The ground-state selection in type-I fcc antiferromagnets was investigated within the second-order perturbation theory used previously in the same context by Lindgård[5,6,20] and Long.[7] We showed that this approach gives results in close agreement with those obtained from the linear spin-wave theory. In agreement with the earlier works,[5,6,20] we found that the ground state is predominantly determined by the quantum effects connected to the strongest spin-spin bonds. Although the predictions of the linear spin-wave theory can be expected to be more accurate, the perturbative calculation is useful as it is simpler and provides analytical results and gives additional insight to the ground-state problem.

We first investigated the case of type-I antiferromagnets with isotropic spin-spin interactions. Two antiferromagnetic structures, separated by a first-order transition, were found as a function of the field: a single-**k** state when $B < 0.407B_c$ and a triple-**k** configuration in $0.407B_c < B < B_c$. The evolution of the triple-**k** structure with field is similar to that found for two-dimensional triangular-lattice antiferromagnets.[26–30] The triple-**k** state assumes the particularly simple up-up-up-down form at $B = B_c/2$. We expect that this structure is stable over a large region of the (T,B) phase diagram in analogy with the up-up-down state of the planar triangular antiferromagnets. Quantum effects tend to stabilize the triple-**k** structure at fields $0.407B_c < B < B_c$ also in weakly anisotropic type-I antiferromagnets, especially for small S.

For type-I fcc antiferromagnets with exchange and dipolar interactions, the ground state was investigated in detail in magnetic fields aligned along the [001] or [110] crystalline direction. Our results are in a fairly good agreement with the spin-wave analysis by Viertiö and Oja,[4] except for a triple-**k** structure at $\mathbf{B} \parallel [001]$ not investigated in their work. The agreement with the earlier perturbation calculations[5,6] was partial.

Our results and the spin-wave calculation[4] identify the high-field antiferromagnetic order of nuclear spins copper as a double-**k** structure when $\mathbf{B} \parallel [001]$ and as a triple-**k** state when $\mathbf{B} \parallel [110]$. These results, as well as the Monte Carlo simulations in Refs. 9, 34, are in agreement with the neutron-diffraction measurements[19] unlike the previous perturbation calculations[5,6] and the Monte Carlo simulations in Ref. 8.

For nuclear ordering in silver, we predict a transition from a single-**k** state to a triple-**k** structure with increasing field for both the [001] and [110] field directions.

Apart from type-I order, we also investigated whether the experimentally observed $\mathbf{k} = (\pi/a)(2/3,2/3,0)$ spin structure in copper could be stabilized by quantum fluctuations. In contrast to earlier work,[5,6,20] we find that quantum effects favor type-I order rather than the $\mathbf{k} = (\pi/a)(2/3,2/3,0)$ configuration. Band-structure calculations of the exchange interactions show, however, that type-I order should be slightly more favorable.[32,33] Our result should thus provide further impetus for refining the calculations of exchange forces in copper.

When $\mathbf{B} \parallel [111]$, no type-I Bragg reflection has been

observed in the high-field phase of the antiferromagnetic region in copper.[19] A solution to this dilemma has been proposed recently by using the mean-field theory[36,48] and the soft-mode approach within the linear spin-wave theory.[49,50] As there is obviously a delicate balance between different antiferromagnetic structures in copper, quantum effects might also be important in the ground-state selection for $\mathbf{B} \parallel [111]$. This problem would provide an interesting extension of this work.

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APPENDIX A: CONNECTION WITH THE SPIN-WAVE (SW) ANALYSIS

The relation of the standard spin-wave result and Eq. (10) given by the perturbation theory can be clarified by solving explicitly the spin-wave ground-state energy and expanding the result in terms of the nondiagonal perturbation. For simplicity we consider at zero field a two-sublattice antiferromagnet with the ordering vector $(\pi/a)(0,0,1)$. The dipolar interaction is neglected and an exchange coupling is assumed between the NN pairs only. In the spin-wave approximation the Hamiltonian is given by

$$H = (H_0 + t\Delta H^{\mathrm{SW}})|_{t=1} \ , \tag{A1}$$

where t is a formal expansion parameter; the higher-order terms are ignored. Introducing Fourier transform and diagonalizing the Hamiltonian we write the quantum correction for the classical ground state energy E_0^{cl} as

$$\delta E_0^{\mathrm{SW}} = 4J_1 S\Big\{-\frac{N}{2} + \sum_{\mathbf{k}}\big[(1 + t\cos k_x a \cos k_y a)^2 - t^2 \cos^2 k_z a(\cos k_x a + \cos k_y a)^2\big]^{\frac{1}{2}}\Big\}\Big|_{t=1} . \tag{A2}$$

The $\mathbf{k}$ sum is to be carried out over a region equivalent to the first Brillouin zone of a sublattice. We find $\delta E_0^{\mathrm{SW}} \approx -0.488 N J_1 S$, in agreement with earlier results.[2,3] Expanding the radical in Eq. (A2) with respect to t and integrating over $\mathbf{k}$ we obtain

$$\delta E_0^{\mathrm{SW}} = J_1 N S\Big(-\frac{1}{2}t^2 + \frac{1}{4}t^3 - \frac{51}{128}t^4 + \frac{9}{16}t^5 - \frac{259}{256}t^6 + \frac{3875}{2048}t^7 - \cdots\Big)\Big|_{t=1} , \tag{A3}$$

and one can identify the coefficient of t^n as the correction δE_n given by the real-space perturbation theory for the Hamiltonian of Eq. (A1). The convergence of expansion (A3) at $t = 1$ is poor and therefore one should not push the perturbative calculation beyond the leading term without a sophisticated summing technique. The poor convergence can be attributed partly to the intrinsic frustration of the fcc lattice. On the other hand, the first term $(-\frac{1}{2})$ alone approximates well the infinite sum (-0.488). Although this may be partly fortuitous, this suggests that rather than using the standard spin-wave theory for calculating ground-state energies one can as well study the same problem in a simpler and more effective way by using the second-order perturbation theory, as has been emphasized by Lindgård[5,6,20] in particular and by Long.[7]

APPENDIX B: EARLIER STUDIES

1. Lindgård's calculation (Ref. 5)

Our results can be compared directly with the work of Lindgård.[5] One should note, however, that in Lindgård's paper the vertical axis is $3D_1/(4J_1+D_1)$ rather than our $d = 3D_1/(J_1 + D_1)$.

When $\mathbf{B} \parallel [001]$, our result agrees with Lindgård's diagram[5] exactly along the lines $d = 0$ and $m = 0$ and quite well for $m < 0.8$. In high fields, $m > 0.8$, there are considerable differences. The most important difference is that Lindgård predicts a second triple-**k** phase with a pattern $\begin{smallmatrix}\downarrow & \circ\\ \uparrow & \circ\end{smallmatrix}$ to appear in a large region at high fields. As we have noticed, together with Viertiö,[51] the $\begin{smallmatrix}\downarrow & \circ\\ \uparrow & \circ\end{smallmatrix}$ structure violates the conditions set by the dipolar interaction, $\mathbf{d}_j \cdot \mathbf{k}_j = 0$, Eq. (4), which requires that the antiferromagnetic modulation must be perpendicular to the respective ordering vector. This clearly increases the energy for the $\begin{smallmatrix}\downarrow & \circ\\ \uparrow & \circ\end{smallmatrix}$ state and the structure cannot be stable.[52] The spins in his high-field single-**k** state for small d are rotated by 45° with respect to the spins in our structure **1a**. In lower fields, our phase lines **1a** ↔ **3** and **2** ↔ **3** differ slightly from those of Lindgård. A possible explanation for this is that his stable triple-**k** structure always appears to be the symmetric triple-**k** state, in which spins in two sublattices are exactly parallel to the field, whereas our stable triple-**k** state acquires the symmetric $\begin{smallmatrix}\leftrightarrows & \circ\\ & \circ\end{smallmatrix}$ form only for $d = 0$, $d = 1$, and $m = 1/2$.

In our and Lindgård's results for $\mathbf{B} \parallel [110]$ (Fig. 5 and the diagram in Ref. 6) the boundaries between the **1z** and **1y** phases are in agreement. There are no triple-**k** structures in his diagram. At high values of d, a double-**k** structure **2yz** mixing **1z** and **1y** like modulations appear: we find **2yz** unstable. Thus most parts of our diagram, Fig. 5, differ from those of Lindgård. We note also that his calculations for $\mathbf{B} \parallel [001]$[5] and $\mathbf{B} \parallel [110]$[6,20] contradict each other because the spin structures at $B = 0$ are not the same.

2. Viertiö-Oja (VO) calculation (Ref. 4)

The standard linear spin-wave theory was used in this study. The spin-spin forces were described by the free-

electron form of the Ruderman-Kittel exchange and by the dipolar force. These interactions were included completely without truncation at nearest neighbors: Differences between the VO model and the present one are thus relatively large. We have examined the VO calculation in relation to our present work by directly comparing the results for the same relative nearest-neighbor interactions J_1/D_1.

a. B ∥ [001]

There is good agreement between the zero-field ground states. VO find for $d < 1.81$, corresponding to their exchange parameter $\eta = 0.94$, the structure **1a**, and for $d > 1.81$ the state **2**.

The field effects were investigated in Ref. 4 for three different ratios of exchange and dipolar interactions. For $d = 2.00$ ($\eta = 0.71$), phase **2** was found to be stable in all fields $B < B_c$. (Our phase **2** and the VO structures P^{β}_{xy} and P^{γ}_{xy} are identical for **B** ∥ [001].)

For $d = 1.79$ ($\eta = 0.96$), VO find two phases as a function of B: structure **1a** [labeled $T(a/2, a/2, 0)$ in Ref. 4] when $m \lesssim 0.1$, and phase **2** when $m \gtrsim 0.1$. Our diagram would give the same result for $d = 1.96$.

Finally, in the limit of a clearly dominant exchange interaction, $d = 0.083$ ($\eta = 50$), VO found that the **1a** phase is stable in all fields $B < B_c$. The absence of structure **3** in this case is the major difference between the two calculations. In fact, structure **3** was not included in the VO ansatz for the ground-state spin configuration for exchange parameters other than those appropriate to copper.[4,53]

b. B ∥ [110]

VO imposed a symmetry ansatz requiring $d_{1y} = d_{2x}$, $d_{1z} = d_{2z}$, $d_{3x} = d_{3y}$ for the triple-**k** structures. The configuration **3a** obeys these conditions but the more general configuration **3b** does not. VO denoted **3a** by P^{α}_{xy} when $m < 1/2$ and by P^{γ}_{xy} when $m > 1/2$. For $d = 2.00$, corresponding to their exchange parameter $\eta = 0.71$, VO find the structure **3a** when $m > 0.23$. When $m < 0.23$, VO obtained a different structure which they denoted by P^{β}_{xy}. This structure does not belong to the **3b** configurations but is obtained from the solution of Eq. (5) for $d_{2x} \neq 0$ ("the second branch"; see text). For $d = 1.79$ ($\eta = 0.96$), VO obtain the phase **1z** when $m < 0.14$, P^{β}_{xy} when $0.14 < m < 0.23$, and **3a** when $m > 0.23$. For $d = 0.083$ ($\eta = 50$), VO find the structure **1y** when $m < 0.55$ and **3a** when m is larger.

3. Studies of thermal fluctuations

Selection of the ground state in type-I fcc antiferromagnets through *thermal* fluctuations has been studied extensively.[8–12] It is not clear whether thermal and quantum effects work in parallel when the interactions are anisotropic, as they do when the interactions are isotropic.[25] Nevertheless, when we compare our results with the Monte Carlo simulations[8–10] of nuclear spin ordering in copper and silver we find a rather good overall agreement. For example, Frisken and Miller[8] obtained the phases **1a** → **3** → **1a** with increasing field along [001] by using a Hamiltonian corresponding to our parameter $d = 1.59$, in a reasonable agreement with Fig. 3. In low intermediate fields, the triple-**k** structure of Frisken and Miller corresponds to our nonsymmetric structure of Eq. (15d) (their phase AF2) whereas in high intermediate fields it is the symmetric triple-**k** state ↰ ° ° (AF3).

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