Antiferromagnets with anisotropic spin-spin interactions: Stability of the zero-field structure in an external field

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The transition from the polarized paramagnetic state to the antiferromagnetic phase in an applied external magnetic field has been investigated theoretically by linear spin-wave theory at $T = 0$. Our analysis applies to spins arranged in a lattice of cubic symmetry. In addition to the Zeeman term, the Hamiltonian consists of bilinear interactions. Antiferromagnetic transition to a state described by an ordering vector $\mathbf{Q}$ is discussed in terms of softening of the corresponding spin-wave excitation in the paramagnetic phase. It is shown that the onset of antiferromagnetic order can be calculated by solving an eigenvalue problem. The smallest eigenvalue of the Fourier transformed 2×2 interaction matrix, which describes the spin-spin interactions in the plane perpendicular to $\mathbf{B}$, determines B_c, $\mathbf{Q}$, and the direction of the antiferromagnetic component. For sufficiently anisotropic spin-spin interactions, these quantities can depend on the direction of $\mathbf{B}$ with respect to the crystalline axes. However, when the spin structure shows an easy-plane anisotropy, which is possible for ordering vectors of the type $\mathbf{Q} = (h, 0, 0)$ and $\mathbf{Q} = (h, h, h)$, and for some vectors at the Brillouin-zone boundary, the direction of $\mathbf{B}$ has no such effect. The general results were first applied to investigate the stability of the easy-axis type-III antiferromagnetism of the fcc lattice, characterized by $\mathbf{Q} = \pi/a(1, 1/2, 0)$. It was shown that, if the spin-spin interactions are sufficiently anisotropic, type-III order becomes unstable against type-I order [$\mathbf{Q} = \pi/a(1, 0, 0)$] when a strong enough field $\mathbf{B}$ is applied along a [111] crystalline axis. If the anisotropy is comparable to the isotropic next-nearest-neighbor coupling, like in K_2IrCl_6, a high-field ordering vector, between the type-I and -III vectors, is predicted. As another application, the magnetic phase diagram of nuclear spins in copper was investigated. Antiferromagnetic type-I ordering has been found in this fcc metal below $T_N = 60$ nK. We studied the puzzle presented by the neutron-diffraction measurements of Annila *et al.*, which show that type-I order is absent in the high-field region below $B_c = 0.25$ mT when $\mathbf{B} \parallel [111]$, although this kind of ordering was observed in the same fields when $\mathbf{B} \parallel [100]$ or [110]. Soft-mode analysis shows that the high-field ordering vector for $\mathbf{B} \parallel [111]$ is of the general type, $\mathbf{Q} = (h, k, l)$, where $|h|$, $|k|$, and $|l|$ are all unequal and nonzero, in agreement with our previous suggestion based on the mean-field theory. We predict various (h, k, l) structures in fields $B \lesssim B_c$ for several field alignments other than [111]. The magnetic phase diagram of nuclear spins in copper can be explained on the basis of the previously calculated spin-spin interactions at least in fields $B \lesssim B_c$ if the calculated parameters are changed only slightly.

I. INTRODUCTION

Anisotropic spin-spin interactions have profound consequences on the ordered spin structure of an antiferromagnet: Since the interactions are not invariant under rotation, the spin directions are coupled to the lattice symmetry. Application of an external magnetic field introduces an additional symmetry-breaking field. It is of interest to study how the ordered spin structure compromises between these two different kinds of energy terms. We address this question by investigating theoretically the transition to the antiferromagnetic phase from the polarized paramagnetic state in the critical field B_c at $T = 0$. It is under these conditions that the possible competition between the magnetic and exchange energies is strongest.

Antiferromagnets are often described by isotropic spin Hamiltonians even though the true interactions contain anisotropy. An isotropic model is easier to treat mathematically and can often account for the property of interest, such as the experimentally determined magnetic structure. When a consistent theoretical picture of the magnetic behavior is desired, however, it is necessary to consider anisotropic spin-spin interactions as well. There is some anisotropy present in all magnetic materials owing to the dipolar force. Although this term is, in most electronic magnets, significantly weaker than the exchange interaction, there are some systems, like cerous magnesium nitrate, in which the dipolar energy dominates. More often, however, the largest anisotropic coupling arises from various exchange mechanisms. The first experiment in which the anisotropy of the exchange interactions was determined is by Griffiths *et al.*[1] Their paramagnetic resonance measurements revealed a significant anisotropic contribution to the spin-spin interactions in K_2IrCl_6 and in $(NH_4)_2IrCl_6$. Later, strongly anisotropic spin-spin interactions have been found in several antiferromagnets, such as various lanthanide and ac-

tinide monopnictides.[2]

Theoretical studies of antiferromagnets with anisotropic interactions were made by ter Haar and Lines.[3,4] They considered the spin structure only at $B = 0$, perhaps because interest in field effects arose only later. Experimental studies on the effect of an external field on the magnetic structure are often complicated by the fact that the routinely accessible fields are limited to 5–10 T, which, with the magnitude of magnetic moment of atoms, corresponds to a thermal energy of 10 K. This is significantly smaller than the nearest-neighbor interaction in most antiferromagnets investigated so far. By now, however, several materials have been found in which the whole field-region for antiferromagnetism is experimentally accessible.[5]

Among the more unconventional systems, strongly anisotropic spin-spin interactions are often encountered in *nuclear* magnets.[6–8] In these spin assemblies the whole field region for antiferromagnetism is accessible because the relevant fields are on the order of 10^{-3} T. The experimental challenge with nuclear magnets is the extremely low ordering temperatures, which are in the nanokelvin region. In nuclear spin assemblies the anisotropic coupling arises both from the dipolar force and from the various exchange mechanisms.[9,10] Analogously with paramagnetic-resonance measurements in electronic magnets, quantitative information about the magnitudes of the isotropic and anisotropic interactions can be obtained in nuclear spin systems by NMR experiments.[11–19] Unlike electronic magnets, exchange forces in nuclear systems can be determined quite reliably from theoretical *ab initio* calculations,[20–24] which are even believed to be 10% accurate in light elements such as copper.

In this paper we discuss antiferromagnetic ordering at $T = 0$ in the field $B = B_c$ by the soft-mode approach. In this technique, the energy needed to excite a spin wave at a wave vector $\mathbf{k}$ in the paramagnetic state is calculated. When a spin wave $\mathbf{k} = \mathbf{Q}$ becomes soft, i.e., when its excitation energy vanishes, the paramagnetic state becomes unstable with respect to antiferromagnetic order, characterized by a spin modulation of wave vector $\mathbf{Q}$, and a phase transition occurs. Our results apply to cubic systems with a Hamiltonian consisting of bilinear interactions, in addition to the Zeeman energy. The type of anisotropy that we treat is due to bilinear spin-spin interactions. We neglect, in particular, single-ion anisotropy due to crystalline fields. This is permissible in spin $S = 1/2$ systems, for which single-ion anisotropy strictly vanishes, and in materials for which the dominating anisotropy is caused by spin-spin interactions.

We apply our results to study the stability of the zero-field structures of several antiferromagnets against other types of antiferromagnetic states in an external field. This problem can be investigated effectively, although somewhat indirectly, by calculating the zero-field structure by linearizing the mean-field equations at $T = T_c$ and by comparing the result to that obtained from linearized equations at $B = B_c(T = 0)$. The calculations are technically simple because Fourier transformations reduce the labor to comparing eigenvalues and eigenvectors of the resulting 3×3 and 2×2 matrices, respectively. The reduction in the dimensions for the latter case reflects the fact that antiferromagnetic order develops, in leading order, only in the plane perpendicular to the field. Although the zero-field transition at $T = T_c$ has been extensively investigated with the eigenvalue analysis, apparently this technique has not been applied to studies of spin ordering at $B = B_c(T = 0)$.

Our work was mostly inspired by the recent studies of nuclear magnetism in Cu.[8,25,26] Additional motivation was given by the observation of nuclear magnetic ordering in Ag,[8,27–29] $PrNi_5$,[30] and Pr,[31] and the search for ordering in Tl,[32,33] Sc,[34,35] Au,[36] $AuIn_2$,[37] and in Rh.[38] The specific problem that we discuss is the phase diagram of nuclear spins in copper, which shows remarkable complexity. Recent neutron diffraction experiments revealed two different magnetic Bragg peaks in the antiferromagnetically ordered structure: (1 0 0) and (0 2/3 2/3) superlattice reflections were found.[26] The relative intensities of the two Bragg peaks depended sensitively on the strength and direction of the external magnetic field.[26,39] At low B, the two reflections appeared simultaneously. This effect, as well as the relative intensities of the two neutron peaks were accounted for by our theoretical model.[40] We also predicted[40,41] the selection rules, prescribing which of the twelve (0 2/3 2/3) reflections, equivalent under cubic symmetry, should appear for the different high-symmetry-field directions. Our rules were verified by later neutron-diffraction measurements.[39] Unexpectedly, however, at fields $B = 0.17 - 0.25$ mT, applied in the [111] direction, neither the (1 0 0) nor the (0 2/3 2/3) Bragg peak was observed, although simultaneous susceptibility measurements indicated antiferromagnetism.

The unknown spin structure for $B \parallel (1,1,1)$ has recently been discussed theoretically by Lindgård[42] and by us.[43] While we used the mean-field theory, Lindgård employed the soft-mode approach. The two calculations yielded dissimilar results for the antiferromagnetic structure. To understand the underlying differences between these two, mainly numerical calculations, we have analytically studied both approaches and their relations with each other. Our present work shows that the mean-field (MF) and soft-mode theories yield, in fact, identical results and confirms our previous conclusions.[43]

The present paper is organized as follows: In Sec. II we first find the spin-wave excitation energies in the paramagnetic state by following the early work by Holstein and Primakoff.[44] We then study the transition between the paramagnetic and antiferromagnetic phases at $T = 0$ by using the soft-mode approach. The critical field for the transition and the wave vector of the soft mode are obtained. The transition is further investigated in Sec. III for various directions of the magnetic field and of the soft-mode wave vectors, which are possible under cubic symmetry. In Sec. IV we briefly compare the soft-mode and mean-field theories of the antiferromagnetic transition at $B_c(T = 0)$. In Sec. V we apply the soft-mode approach first to investigate the stability of type-III ordering in an fcc lattice at $B \neq 0$. We then discuss the

magnetic phase diagram of nuclear spins in copper and make some comments on the corresponding diagrams of silver and gold. The main results of our paper are summarized in Sec. VI.

II. SOFT MODES

A. Spin-wave energies

The Hamiltonians of nuclear spins in copper and silver and of several other spin assemblies in a cubic crystal consist of bilinear spin-spin interactions and the Zeeman energy, viz.,

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

where $\mathbf{S}_i$ is the spin operator acting at site i, and the 3×3 matrix $\underline{A}_{ij}$ describes the interaction between spins i and j. $\mathbf{B}$ is the applied external magnetic field. The coefficient $\hbar\gamma$, where γ is the gyromagnetic ratio, has been absorbed in $\mathbf{B}$ so that the field has the units of energy. $\underline{A}_{ij}$ consists of the dipolar and exchange forces; the latter interaction may be isotropic[9] or anisotropic.[10] The symmetry of the anisotropic exchange coupling need not be dipole-like but can be of more general form.[23]

Constraints on the form of $\underline{A}_{ij}$ are imposed by the symmetry of the lattice, which is here assumed to be cubic. First, because of the inversion symmetry of the crystal, $\underline{A}_{ij}$ is a real and symmetric matrix. If the crystal did not display inversion symmetry, an antisymmetric, Dzyaloshinski-Moriya type of interaction would be possible as well. Second, $\underline{A}_{ij}$ transforms like a second rank tensor with respect to $\mathbf{r}_{ij}$, the vector joining spins i and j. This imposes certain conditions for the elements of $\underline{A}_{ij}$ when $\mathbf{r}_{ij}$ lies in a symmetry direction of the crystal. For example, if $\mathbf{r}_{ij}$ is of the form $(\ell,\ell,0)$, the components of $\underline{A}_{ij}$ satisfy $A^{xx}_{ij} = A^{yy}_{ij}$ and $A^{xz}_{ij} = A^{yz}_{ij} = 0$. There are altogether six different cases for $\mathbf{r}_{ij}$ in a cubic lattice; the corresponding forms of $\underline{A}_{ij}$ have been listed in Ref. 23.

The spin-wave energies for the Hamiltonian of Eq. (1) have been derived by Holstein and Primakoff.[44] The scheme of their calculation, which is now familiar from several textbooks,[45] is the following: The Hamiltonian is rewritten using the spin-deviation operators, higher-order terms than those bilinear in the spin-deviation operators are neglected, the resulting Hamiltonian is Fourier transformed and finally diagonalized by use of the Bogoliubov transformation. This procedure yields the normal modes, i.e., spin waves, and the corresponding energies. The excitation energy for a spin wave is given by

$$\omega_{\mathbf{k}} = \sqrt{C^2_{\mathbf{k}} - 4|D_{\mathbf{k}}|^2} \ , \tag{2}$$

where

$$\begin{aligned} C_{\mathbf{k}} &= \frac{S}{2}[A^{xx}(\mathbf{k}) + A^{yy}(\mathbf{k})] + B - SA^{zz}(0) \ , \\ D_{\mathbf{k}} &= \frac{S}{4}[A^{xx}(\mathbf{k}) - A^{yy}(\mathbf{k}) - 2iA^{xy}(\mathbf{k})] \ . \end{aligned} \tag{3}$$

Here $\underline{A}(\mathbf{k})$ is the Fourier transform of the interaction matrix, viz.,

$$A^{\alpha\beta}(\mathbf{k}) = \sum_j A^{\alpha\beta}_{ij} e^{i\mathbf{k}\cdot\mathbf{r}_{ij}} \ . \tag{4}$$

It is clear from Eq. (2) that in a high enough field all excitation energies $\omega_{\mathbf{k}}$ are positive. With decreasing field one of the spin-wave energies eventually becomes negative indicating softening of this particular excitation and instability of the paramagnetic phase.

B. Isotropic interactions

It is instructive to consider first isotropic interactions, $A^{\alpha\beta}_{ij} = J_{ij}\delta^{\alpha\beta}$. The spin-wave energies are then

$$\omega_{\mathbf{k}} = B + S[J(\mathbf{k}) - J(0)] \ , \tag{5}$$

where $J(\mathbf{k})$ is the Fourier transform of J_{ij}. When the field is lowered, the soft mode appears for the wave vector $\mathbf{Q}$, which corresponds to the minimum of $J(\mathbf{k})$: $J(\mathbf{Q}) = \min_{\mathbf{k}}\{J(\mathbf{k})\}$. The critical field B_c for the soft-mode transition is

$$B_c = S[J(0) - J(\mathbf{Q})] \ , \tag{6}$$

and the transition is continuous. B_c and $\mathbf{Q}$ are independent of the orientation of $\mathbf{B}$.

The ordered spin configuration below B_c is beyond the applicability of the spin-wave theory for the paramagnetic state. The natural interpretation for the appearance of a soft mode at a wave vector $\mathbf{Q}$ is, however, that immediately below B_c there is a small antiferromagnetic component, described by $\mathbf{Q}$, in the plane $\perp \mathbf{B}$.

C. Anisotropic interactions

To study the soft-mode transition in the presence of anisotropic interactions, we rewrite the spin-wave energy of Eq. (2) in the form

$$\omega^2_{\mathbf{k}}/S^2 = \det\begin{pmatrix} A^{xx}(\mathbf{k}) + B/S - A^{zz}(0) & A^{xy}(\mathbf{k}) \\ A^{xy}(\mathbf{k}) & A^{yy}(\mathbf{k}) + B/S - A^{zz}(0) \end{pmatrix} \ . \tag{7}$$

The determinant vanishes when $-B/S+A^{zz}(0)$ coincides with an eigenvalue $\tilde{\lambda}(\mathbf{k})$ of

$$\underline{\tilde{A}}(\mathbf{k}) = \begin{pmatrix} A^{xx}(\mathbf{k}) & A^{xy}(\mathbf{k}) \\ A^{xy}(\mathbf{k}) & A^{yy}(\mathbf{k}) \end{pmatrix} . \tag{8}$$

This 2×2 matrix is the xy block of the 3×3 matrix $\underline{A}(\mathbf{k})$ and corresponds to the plane perpendicular to the field direction $\hat{z}$. Thus the soft-mode transition takes place at

$$B_c = S[\lambda(0) - \tilde{\lambda}(\tilde{\mathbf{Q}})] \tag{9}$$

to a state described by the wave vector $\tilde{\mathbf{Q}}$, where $\lambda(0) = A^{zz}(0)$ and

$$\tilde{\lambda}(\tilde{\mathbf{Q}}) = \min_{\mathbf{k},i}\{\tilde{\lambda}_i(\mathbf{k})\} . \tag{10}$$

The matrix $\underline{\tilde{A}}(\mathbf{k})$ clearly depends on the direction of the external field. Therefore, the eigenvalues $\tilde{\lambda}_i(\mathbf{k})$ can also depend on the direction of $\mathbf{B}$, and so can the ordering vector $\tilde{\mathbf{Q}}$ and the critical field B_c.

To find the relationship between the two eigenvalues of $\underline{\tilde{A}}(\mathbf{k})$ and the three eigenvalues of $\underline{A}(\mathbf{k})$, we write $\underline{A}(\mathbf{k})$ in terms of its eigenvalues λ_i and the normalized eigenvectors $\hat{\mathbf{e}}_i$, $i = 1, 2, 3$, viz.,

$$\underline{A}(\mathbf{k}) = \sum_{i=1}^{3} \lambda_i \hat{\mathbf{e}}_i \hat{\mathbf{e}}_i . \tag{11}$$

When rotated to an orthogonal basis $\{\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}}\}$, the matrix $\underline{A}(\mathbf{k})$ becomes

$$\begin{aligned} \underline{A}(\mathbf{k}) &\rightarrow \sum_{\alpha=x,y,z} \hat{\boldsymbol{\alpha}}\hat{\boldsymbol{\alpha}} \cdot \sum_{i=1}^{3} \lambda_i \hat{\mathbf{e}}_i \hat{\mathbf{e}}_i \cdot \sum_{\beta=x,y,z} \hat{\boldsymbol{\beta}}\hat{\boldsymbol{\beta}} \\ &= \sum_{\alpha\beta} \hat{\boldsymbol{\alpha}}\hat{\boldsymbol{\beta}} \left(\sum_i \lambda_i (\hat{\mathbf{e}}_i \cdot \hat{\boldsymbol{\alpha}})(\hat{\mathbf{e}}_i \cdot \hat{\boldsymbol{\beta}}) \right) , \end{aligned} \tag{12}$$

where α and β run over x, y, and z. Denoting the field direction by $\hat{\mathbf{z}}$, the xy block equals $\underline{\tilde{A}}(\mathbf{k})$. Let $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ be the two eigenvectors of $\underline{\tilde{A}}(\mathbf{k})$. The xy block is then diagonal and the eigenvalues of $\underline{\tilde{A}}(\mathbf{k})$ are

$$\tilde{\lambda}_1(\mathbf{k}) = \sum_{i=1}^{3} \lambda_i (\hat{\mathbf{e}}_i \cdot \hat{\mathbf{x}})^2 \tag{13}$$

and

$$\tilde{\lambda}_2(\mathbf{k}) = \sum_{i=1}^{3} \lambda_i (\hat{\mathbf{e}}_i \cdot \hat{\mathbf{y}})^2 . \tag{14}$$

Since $\sum_i (\hat{\mathbf{e}}_i \cdot \hat{\mathbf{x}})^2 = \sum_i (\hat{\mathbf{e}}_i \cdot \hat{\mathbf{y}})^2 = 1$, the two eigenvalues of $\underline{\tilde{A}}(\mathbf{k})$ are bounded by the three eigenvalues of $\underline{A}(\mathbf{k})$, i.e.,

$$\min_i\{\lambda_i(\mathbf{k})\} \le \tilde{\lambda}_j(\mathbf{k}) \le \max_\ell\{\lambda_\ell(\mathbf{k})\} . \tag{15}$$

The relationship between the eigenvalues $\lambda_j(\mathbf{k})$ and $\tilde{\lambda}_j(\mathbf{k})$ is visualized by Fig. 1. When a real and symmetric 3×3 matrix operates on the unit vectors of a sphere, an ellipsoid is created. The lengths of the principal axes are given by the eigenvalues λ_i, which can be taken as positive numbers for the illustration. The intersection of the ellipsoid and the plane perpendicular to the field direction $\hat{\mathbf{z}}$ defines an ellipse, the principal axes of which correspond to the eigenvalues $\tilde{\lambda}_i$ of the 2×2 matrix $\underline{\tilde{A}}(\mathbf{k})$. It is obvious that the lengths of the principal axes of this ellipse are bounded from above and below by those of the ellipsoid, in accordance to Eq. (15).

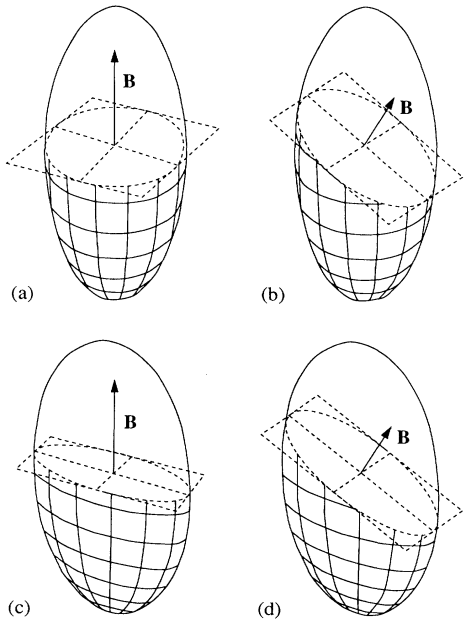

FIG. 1. Illustration of the relationships between the eigenvalues of the real and symmetric 3×3 matrix $\underline{A}$ and its 2×2 submatrix $\underline{\tilde{A}}$. The three eigenvalues λ_i ($\lambda_1 \le \lambda_2 \le \lambda_3$) of $\underline{A}$ describe the lengths of the principal axes of the ellipsoid, while the two eigenvalues $\tilde{\lambda}_i$ of $\underline{\tilde{A}}$ are equal to the principal axes of the ellipse drawn using the broken line. The ellipse is determined as the intersection of the ellipsoid and a plane perpendicular to the external field $\mathbf{B}$. (a) and (b): Degenerate eigenvalues $\lambda_1 = \lambda_2$. In (a), $\mathbf{B}$ is $\perp$ to the plane spanned by $\mathbf{e}_1$ and $\mathbf{e}_2$, leading to $\tilde{\lambda}_1 = \tilde{\lambda}_2 = \lambda_{1,2}$. In (b), $\mathbf{B}$ is in a general direction, leading to $\tilde{\lambda}_1 = \lambda_{1,2} \le \tilde{\lambda}_2$. (c) and (d): Nondegenerate eigenvalues λ_i. In (c), $\mathbf{B}$ is $\perp$ to the plane spanned by $\mathbf{e}_1$ and $\mathbf{e}_2$, leading to $\tilde{\lambda}_1 = \lambda_1$ and $\tilde{\lambda}_2 = \lambda_2$. In (d), $\mathbf{B}$ has a general alignment, leading to $\lambda_1 \le \tilde{\lambda}_1$ and $\tilde{\lambda}_2 \le \lambda_3$.

An important special case for which two of the eigenvalues of $\underline{A}(\mathbf{k})$ are degenerate is illustrated in Figs. 1(a) and 1(b). For any field direction, the ellipsoid and the ellipse have at least one common principal axis, i.e., eigenvalue. If the lowest eigenvalue $\lambda_{\min}$ of $\underline{A}(\mathbf{k})$ is degenerate, it follows that the soft-mode transition always takes place for the corresponding wave vector $\mathbf{Q}$ at the same value of the critical field, independent of its direction.

In the general case, the eigenvalues of $\underline{A}(\mathbf{k})$ are nondegenerate, as illustrated in Figs. 1(c) and 1(d). If this is the case for the zero-field ordering vector, which corresponds to the eigenvalue $\lambda_{\min}$, an intersection by a plane perpendicular to $\mathbf{B}$ yields, in general, eigenvalues $\tilde{\lambda}_i$ which are larger than $\lambda_{\min}$ [Fig. 1(d)]. The eigenvalues are equal only when the field is exactly perpendicular to

the principal axis corresponding to $\lambda_{\min}$. It is then possible that a vector $\tilde{\mathbf{Q}}$, different from the zero-field ordering vector $\mathbf{Q}$, yields $\min_{i,\mathbf{k}}\{\tilde{\lambda}_i(\mathbf{k})\}$. Then the wave vector of the soft mode and the critical field can depend on the direction of $\mathbf{B}$.

If the ordering vectors in zero field at $T = T_c$ and at $B = B_c$ when $T = 0$ are different, the phase boundary between the paramagnetic and antiferromagnetic states in the B-T plane should show an interesting cross-over between these two limiting cases. This effect will be investigated in a forthcoming paper.[46]

III. SOFT-MODE TRANSITIONS FOR DIFFERENT FIELD ALIGNMENTS

A. Classification of the antiferromagnetic modes perpendicular to the field

Let us see what we can conclude about the antiferromagnetic transition at $B = B_c$ by knowing only the wave vector $\mathbf{Q}$, which yields the lowest eigenvalue $\lambda_{\min}$ of the Fourier transformed 3×3 interaction matrix $\underline{A}(\mathbf{k})$. Our analysis in Sec. II C showed that if $\lambda_{\min}$ is degenerate, the ordering vectors at zero field when $T = T_c$ and at $B = B_c$ when $T = 0$ are the same. In this section we find the ordering vectors in cubic systems for which this simple behavior occurs. It turns out, however, that the eigenvalues are nondegenerate for the most zero-field ordering vectors $\mathbf{Q}$. Therefore, the wave vector for the soft-mode transition into $\tilde{\mathbf{Q}}$ is, in general, different from $\mathbf{Q}$. However, if the field is applied in a cubic high-symmetry direction, it is possible that $\tilde{\mathbf{Q}} = \mathbf{Q}$, depending on the symmetry. In fact, in several cases it is easy to show that $\tilde{\mathbf{Q}} = \mathbf{Q}$ even if the particular spin-spin interactions are not known, but in the remaining cases one does not know whether $\tilde{\mathbf{Q}} \neq \mathbf{Q}$ without calculations using the actual exchange parameters.

Among the ordering vectors $\mathbf{Q}$ that we have investigated, those of the form $(h,0,0)$, (h,h,h), $(h,h,0)$ possess high symmetry. We have also studied the less symmetric cases $\mathbf{Q} = (h,k,0)$ and $\mathbf{Q} = (h,h,l)$, and the case with no symmetry, $\mathbf{Q} = (h,k,l)$, where $|h|$, $|k|$, and $|l|$ are all unequal and nonzero. These are the six possibilities in a cubic crystal. Likewise, there are six symmetrically inequivalent directions of the external field. We denote the field alignment somewhat differently to avoid confusion, namely, [100], [111], [110], $[\xi\eta 0]$, $[\xi\xi\eta]$, and $[\xi\eta\zeta]$.

Let us first consider ordering vectors of the form $(h,0,0)$. Then, owing to the cubic symmetry,

$$\underline{A}(h,0,0) = \begin{pmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & b \end{pmatrix} \tag{16}$$

with eigenvalues $\lambda_1 = a$ and $\lambda_2 = \lambda_3 = b$. The respective eigenvectors are $\mathbf{e}_1 = (1,0,0)$ and $\mathbf{e}_2$ and $\mathbf{e}_3$, which span the plane perpendicular to $\mathbf{e}_1$. If $\lambda_{\min}$ equals the degenerate λ_2 and λ_3, the soft mode always appears at $\mathbf{k} = \mathbf{Q}$ [Figs. 1(a) and 1(b)]. The number of soft modes is, however, not the same for different field alignments. For $\mathbf{B} \parallel (1,0,0)$, the two normal modes corresponding to $\mathbf{k} = (h,0,0)$ become soft at B_c because both eigenvectors $\mathbf{e}_2$ and $\mathbf{e}_3$ are $\perp\mathbf{B}$ [Fig. 1(a)]. In addition, there is one soft mode for $\mathbf{k} = (0,h,0)$ and $\mathbf{k} = (0,0,h)$ each [Fig. 1(b)]. Taking also into account the modes at $(-h,0,0)$, $(0,-h,0)$, and $(0,0,-h)$, we find altogether eight soft modes at B_c for the six vectors in the star of $\mathbf{Q}$, which we denote by $\{\mathbf{Q}\}^*$ or, in the component form, by $\langle Q_x, Q_y, Q_z\rangle$. For field alignments $\mathbf{B} \parallel (1,1,1)$ and $\mathbf{B} \parallel (1,1,0)$, it is possible to find one eigenvector $\perp \mathbf{B}$ for each member of $\{\mathbf{Q}\}^*$. There are thus six soft modes at $B = B_c$.

If the number of soft modes depends on the field alignment, as in the case discussed above, it is possible that so does the order of the transition. In fact, we believe that the number of soft modes at $B = B_c$ equals the dimensionality of the order parameter; this is a crucial quantity in the Landau-Ginzburg-Wilson theory of phase transitions.[47]

Let us then consider such an anisotropy that the eigenvalue λ_1 for $\mathbf{Q} = (h,0,0)$ is lowest; the eigenvector $\mathbf{e}_1$ is then parallel to $(1,0,0)$. For $\mathbf{Q} = (0,h,0)$ and $\mathbf{Q} = (0,0,h)$ the respective eigenvectors are $(0,1,0)$ and $(0,0,1)$. If $\mathbf{B}$ is perpendicular to one of these eigenvectors, a soft-mode transition takes place at the corresponding wave vector, e.g., at $\mathbf{k} = (0,\pm h,0)$ and $\mathbf{k} = (0,0,\pm h)$ when $\mathbf{B} \parallel (1,0,0)$, and at $\mathbf{k} = (0,0,\pm h)$ when $\mathbf{B} \parallel (1,1,0)$. For example, if $\mathbf{B} \parallel (1,1,1)$, no eigenvector corresponding to $\langle h,0,0\rangle$ order is perpendicular to the field. The soft-mode transition may or may not take place at a $\langle h,0,0\rangle$ vector. Generally, it is only known that the transition occurs in a field that is lower than $B = S[\lambda(0) - \lambda_1(h,0,0)]$. The wave vector and the field at which the transition takes place depend on the actual values of $A_{ij}^{\alpha\beta}$.

The analysis for ordering vectors of the form (h,h,h) is rather similar to the case $\mathbf{Q} = (h,0,0)$: There is again a unique eigenvalue and two degenerate ones. If $\lambda_{\min}$ equals the degenerate λ_2 and λ_3, there is a soft-mode transition at a $\langle h,h,h\rangle$ vector for an arbitrary field alignment, whereas if $\lambda_{\min} = \lambda_1$, there is a soft-mode transition at a $\langle h,h,h\rangle$ vector, provided that the field is perpendicular to one of the eight eigenvectors $(\pm 1,\pm 1,\pm 1)$. For example, when $\mathbf{B} \parallel (1,1,0)$, magnons with wave vectors $(h,-h,\pm h)$, and $(-h,h,\pm h)$ become simultaneously soft at $B_c = S[\lambda(0) - \lambda_1(h,h,h)]$. When $\mathbf{B} \parallel (1,0,0)$ or $\mathbf{B} \parallel (1,1,1)$ one cannot assess, in general, whether a soft-mode transition takes place at a $\langle h,h,h\rangle$ vector or not.

The common feature of the ordering vectors $\mathbf{Q} = (h,h,0)$, $(h,k,0)$, (h,h,l), and (h,k,l) is that the eigenvalues of $\underline{A}(\mathbf{Q})$ are unique. Therefore, for an arbitrary nonsymmetric alignment of $\mathbf{B}$, it is not known whether a soft-mode transition takes place at one of the vectors of $\{\mathbf{Q}\}^*$. For fields applied along the high-symmetry directions [100], [111], and [110] it is, however, often possible to find an eigenvector of $\underline{A}(\mathbf{Q})$, which is $\perp \mathbf{B}$ and which, therefore, becomes soft at $B = S[\lambda(0) - \lambda_{\min}(\mathbf{Q})]$.

The eigenvectors of $\underline{A}(\mathbf{Q})$ can be deduced if $\mathbf{Q}$ lies in one of the crystalline symmetry directions. Consider, for example, $\mathbf{Q} = (h,h,0)$. The Fourier sum of the interac-

tion matrix $\underline{A}_{ij}$ has the form

$$\underline{A}(h,h,0) = \begin{pmatrix} a & d & 0 \\ d & a & 0 \\ 0 & 0 & c \end{pmatrix} , \tag{17}$$

which has eigenvectors $(1,1,0)$, $(1,-1,0)$, and $(0,0,1)$. When $\mathbf{Q} = (h,k,0)$, $h \neq k$, there are two eigenvectors $\mathbf{e}_1$ and $\mathbf{e}_2$ in the xy plane, while $\mathbf{e}_3 = (0,0,1)$. When $\mathbf{Q} = (h,h,l)$,

$$\underline{A}(h,h,l) = \begin{pmatrix} a & d & e \\ d & a & e \\ e & e & c \end{pmatrix} . \tag{18}$$

One eigenvector is (1,−1,0) and the other two have the forms $(1,1,\alpha)$ and $(\beta,\beta,1)$.

In Table I, we summarize our results of the soft-mode analysis for magnetic structures in a cubic lattice. Wave vectors at zone boundaries deserve, however, special attention and must be treated separately. For example, $\mathbf{k}$ and $-\mathbf{k}$ are equivalent at the zone boundary, whereas they have been counted as two different vectors in Table I. Furthermore, the eigenvectors can have special symmetric directions and the eigenvalues may display additional degeneracy. Consider, for example, the zone-boundary vector in the reciprocal lattice of the body-centered-cubic system, $\mathbf{Q} = \pi/a(1,1,1)$, with $2a$ equal to the lattice constant. $\underline{A}(\mathbf{Q})$ is proportional to the unit matrix so that its three eigenvalues are degenerate. This is a reflection of the fact that the bcc lattice consists of two interpenetrating simple-cubic lattices so that a two-sublattice antiferromagnet does not break the symmetry. In fact, most observed antiferromagnetic structures correspond to zone-boundary wave vectors. For example, of the four conventional fcc antiferromagnets described by ordering vectors $\mathbf{Q} = \pi/a(1,0,0)$ (type I), $\mathbf{Q} = \pi/a(1/2,1/2,1/2)$ (type II), $\mathbf{Q} = \pi/a(1,1/2,0)$ (type III), and $\mathbf{Q} = \pi/a(1/2,1/2,0)$ (type-IV), structures of type I, II, and III correspond to a zone-boundary vector. In Sec. V A we shall discuss the zone-boundary effects on the eigenvalues and eigenvectors of $\underline{A}(\mathbf{Q})$ in type-III fcc antiferromagnets.

B. Stability of the ordering vector Q corresponding to $\lambda_{\min}$

In Sec. III A we listed the cases for which the symmetry requires that the wave vector $\tilde{\mathbf{Q}}$ of the soft-mode transition equals the zero-field ordering vector $\mathbf{Q}$. It was shown that for a nondegenerate $\lambda_{\min}$, this happens only

TABLE I. Number of soft modes for the various wave vectors $\mathbf{Q}$ and for different directions of the external magnetic field $\mathbf{B}$, which are possible under cubic symmetry. The modes become soft in a field $B_c = S[\lambda(0) - \lambda_n(\mathbf{Q})]$, provided that no other mode becomes soft in a higher field. This is equivalent to assuming that the lowest eigenvalue $\lambda_{\min} = \lambda_n(\mathbf{Q})$. The number of soft modes expresses how many of the eigenvectors $\mathbf{e}_n(\mathbf{Q})$ are perpendicular to the field direction for the vectors in the star of $\mathbf{Q}$, $\{\mathbf{Q}\}^*$. When the quoted number is zero, softening of the mode is, nevertheless, possible in the field $B_c = S[\lambda(0) - \lambda_n(\mathbf{Q})]$ if the eigenvector $\mathbf{e}_n(\mathbf{Q})$ is accidentally perpendicular to $\mathbf{B}$, and in lower fields as well if no other mode becomes soft in a higher field.

$\mathbf{Q}$	$(h,0,0)$	(h,h,h)	$(h,h,0)$	$(h,k,0)$	(h,h,l)	(h,k,l)
Members in $\{\mathbf{Q}\}^*$	6	8	12	24	24	48
$\mathbf{e}_1$	$(1,0,0)$	$(1,1,1)$	$(1,1,0)$	$(\alpha,\beta,0)$	$(1,1,\alpha)$	no symmetry
$\mathbf{e}_2$	in plane $\perp \mathbf{e}_1$	in plane $\perp \mathbf{e}_1$	(1,−1,0)	$(\beta,-\alpha,0)$	(1,−1,0)	no symmetry
$\mathbf{e}_3$	in plane $\perp \mathbf{e}_1$	in plane $\perp \mathbf{e}_1$	(0,0,1)	(0,0,1)	$(\beta,\beta,1)$	no symmetry

Number of soft modes

$\min\{\lambda_1,\lambda_2,\lambda_3\}$	λ_1	$\lambda_2=\lambda_3$	λ_1	$\lambda_2=\lambda_3$	λ_1 or λ_2	λ_3	λ_1 or λ_2	λ_3	λ_1 or λ_3	λ_2	Any λ
B along											
[100]	4	8	0	8	4	8	8	16	0	8	0
[111]	0	6	0	10	6	0	0	0	0	12	0
[110]	2	6	4	8	2	4	0	8	4	4	0
$[\xi\eta 0]$	2	6	0	8	0	4	0	8	0	0	0
$[\xi\xi\eta]$	0	6	0	8	2	0	0	0	0	4	0
$[\xi\eta\zeta]$	0	6	0	8	0	0	0	0	0	0	0

for external fields aligned in the planes perpendicular to the eigenvector $\mathbf{e}(\mathbf{k})$, where $\mathbf{k} \in \{\mathbf{Q}\}^*$. In this section we study whether $\tilde{\mathbf{Q}}$ equals $\mathbf{Q}$ even when the field is slightly tilted away from one of these planes. Knowing that $\tilde{\mathbf{Q}} = \mathbf{Q}$ when the interactions are isotropic, we wish to find out whether the strength of the anisotropic interactions needs to be above a certain level for $\tilde{\mathbf{Q}}$ to depart from $\mathbf{Q}$ or whether this is possible for arbitrarily small anisotropic terms. One expects *a priori* the first possibility because experiments seldom show that the ordering vector depends on the field alignment.

Let us first assume that $\tilde{\mathbf{Q}} = \mathbf{Q}$ even when the field is tilted by an angle θ out of the plane $\perp \mathbf{e}(\mathbf{Q})$. This would be seen as a decrease in B_c or, equivalently, as an increase in the eigenvalue $\tilde{\lambda}_{\min}(\mathbf{Q})$. According to Eqs. (13)–(14), the change in $\tilde{\lambda}_{\min}(\mathbf{Q})$ can be as large as

$$\Delta\tilde{\lambda}_{\min}(\mathbf{Q}) = [\lambda_{\max}(\mathbf{Q}) - \lambda_{\min}(\mathbf{Q})]\,\theta^2 \ . \tag{19}$$

The increase $\Delta\tilde{\lambda}_{\min}(\mathbf{Q})$ is clearly proportional to the anisotropy of the interactions.

We then assume that a new ordering vector $\mathbf{Q}'$ close to $\mathbf{Q}$ is stabilized by the tilt because the eigenvector $\mathbf{e}(\mathbf{Q}')$ is more closely perpendicular to the field than $\mathbf{e}(\mathbf{Q})$. The eigenvalue $\lambda_{\min}(\mathbf{Q}')$ is, however, larger than $\lambda_{\min}(\mathbf{Q})$, which tends to destabilize $\mathbf{Q}'$. Let us assume for simplicity that the new eigenvector $\mathbf{e}(\mathbf{Q}')$ is exactly perpendicular to the tilted $\mathbf{B}$ so that the increase in the eigenvalue due to the tilt is $\Delta\lambda = \lambda_{\min}(\mathbf{Q}') - \lambda_{\min}(\mathbf{Q})$. To estimate $\Delta\lambda$ we note that the change in the ordering vector $\Delta\mathbf{k} = \mathbf{Q}' - \mathbf{Q}$ has to be sufficiently large to rotate the eigenvector at least by an angle θ. Since the components of $\mathbf{e}(\mathbf{k})$ transform like the corresponding components of $\mathbf{k}$, we may write $(\Delta\mathbf{e}(\mathbf{k}))_\alpha \propto (\Delta\mathbf{k})^n_\alpha$, where $n = 1, 3, 5, \ldots$. The required change $\Delta\mathbf{k}$ for a fixed θ is smallest when $n = 1$. It then follows that $|\,\Delta\mathbf{k}\,| \approx \theta\,|\,\mathbf{Q}\,|$. Let us further assume that the isotropic term dominates the spin-spin interactions. Since the gradient of $\lambda_{\min}(\mathbf{k})$ vanishes at $\mathbf{k} = \mathbf{Q}$, we obtain

$$\Delta\lambda \approx \frac{1}{2}[(\hat{\mathbf{q}}\cdot\boldsymbol{\nabla}_{\mathbf{k}})^2 J(\mathbf{k})]_{\mathbf{k}=\mathbf{Q}}\,\theta^2 Q^2 \ , \tag{20}$$

where $\hat{\mathbf{q}}$ denotes a unit vector along $\Delta\mathbf{k}$. The contribution from the anisotropic interactions to $\Delta\lambda$ has been neglected. If the increase in the eigenvalue due to a change in the ordering vector, is larger than that due to the anisotropy, i.e., if

$$\frac{1}{2}[(\hat{\mathbf{q}}\cdot\boldsymbol{\nabla}_{\mathbf{k}})^2 J(\mathbf{k})]_{\mathbf{k}=\mathbf{Q}}\,Q^2 \gtrsim \lambda_{\max}(\mathbf{Q}) - \lambda_{\min}(\mathbf{Q}) \ , \tag{21}$$

the ordering vector $\mathbf{Q}$ is stable even though the field is tilted away from the plane $\perp \mathbf{e}(\mathbf{Q})$. Since the left-hand side of Eq. (21) is linearly proportional to the isotropic interaction, the anisotropic force has to be sufficiently large in comparison to its isotropic counterpart to cause a change in the ordering vector when the field is rotated with respect to the crystalline axes.

In Eq. (20) we assumed that the second derivative of $J(\mathbf{k})$ does not vanish. If it were equal to zero, which to us seems very unlikely, the ordering vector would change even for an arbitrarily small anisotropic interaction.

IV. COMPARISON WITH THE MEAN-FIELD THEORY

A. Isotropic interactions

In the MF theory, the ground state is always described by the wave vector $\mathbf{Q}$ that corresponds to the minimum of $J(\mathbf{k})$, i.e., $J(\mathbf{Q}) = \min_{\mathbf{k}}\{J(\mathbf{k})\}$. In an external field, the spin structure of minimum energy for an arbitrary $\mathbf{Q}$ is given by

$$\langle \mathbf{S}_i \rangle = \sqrt{S^2 - m^2}[\hat{\mathbf{x}}\cos(\mathbf{Q}\cdot\mathbf{r}_i) + \hat{\mathbf{y}}\sin(\mathbf{Q}\cdot\mathbf{r}_i)] + m\hat{\mathbf{z}} \ , \tag{22}$$

where

$$m = \frac{B}{J(0) - J(\mathbf{Q})} \ . \tag{23}$$

The structure is conical with the antiferromagnetic component rotating in the plane $\perp \mathbf{B}$. In the field $B_c = S[J(0) - J(\mathbf{Q})]$, the cone transforms continuously into the polarized paramagnetic state. Both B_c and $\mathbf{Q}$ are independent of the field alignment. Therefore, the MF theory for the ordered state and the soft-mode calculation for the paramagnetic phase give a mutually consistent picture.

B. Anisotropic interactions

No general solution for the MF ground state of an antiferromagnet with anisotropic spin-spin interactions has been presented so far, not even for the spin structure immediately below B_c. We have recently studied this question. In a forthcoming paper, we will present an equation for the structure in the vicinity of the phase boundary in the B-T plane.[46] At $B = B_c$ ($T = 0$), the MF result reduces to the soft-mode solution. The ordering vector immediately below B_c is the one that yields the lowest eigenvalue for the matrix $\underline{\tilde{A}}(\mathbf{k})$ of Eq. (16), with $B_c = S[\lambda(0) - \tilde{\lambda}_{\min}]$. Antiferromagnetism at $T = 0$ develops in the plane perpendicular to the external field. Since only the in-plane interactions matter, the order is determined by the corresponding 2×2 block of the 3×3 interaction matrix.

V. APPLICATIONS

A. Stability of type-III ordering in $B \neq 0$

Several antiferromagnets of an fcc crystal lattice show type-III ordering, like K_2IrCl_6,[48,49] β-MnS,[50] and MnS_2.[51] This spin structure is characterized by the six $\mathbf{k}$ vectors belonging to the star of $\mathbf{Q} = (\pi/a)(1, 1/2, 0)$. We show here that antiferromagnets that are of type III at $B = 0$ can undergo phase transitions in an applied field.

We first derive some well-known properties of type-III order using eigenvalue analysis. At the same time we show how the eigenvalues and eigenvectors of the Fourier transform of the interaction matrix are affected when the ordering vector is at the zone boundary. $\underline{A}(\mathbf{k})$ for $\mathbf{k} =$

$(k_x, k_y, 0)$ has the form

$$\underline{A}(k_x, k_y, 0) = \begin{pmatrix} a & d & 0 \\ d & b & 0 \\ 0 & 0 & c \end{pmatrix} . \tag{24}$$

If $k_x = \pi/a$, so that $\mathbf{k}$ is at the boundary of the first Brillouin zone, $\underline{A}(\pi/a, k_y, 0) = \underline{A}(-\pi/a, k_y, 0)$. The xy component of this matrix equation reads $d = -d$, since $\underline{A}^{\alpha\beta}(\mathbf{k})$ transforms like $k_\alpha k_\beta$. Thus $d = 0$ and $\underline{A}(\pi/a, k_y, 0)$ is diagonal.

Since the type-III ordering vector $\pi/a(1, 1/2, 0)$ is a corner point of the first Brillouin zone, it follows also that $\underline{A}[\pi/a(1, 1/2, 0)] = \underline{A}[\pi/a(0, -1/2, -1)]$ yielding $A^{xx} = A^{zz}$, i.e., $a = c$. Thus

$$\underline{A}[\pi/a(1, 1/2, 0)] = \begin{pmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & a \end{pmatrix} . \tag{25}$$

The magnetic unit cell of a $\mathbf{Q} = \pi/a(1, 1/2, 0)$ structure is obtained by doubling the fcc unit cell in the [010] direction as shown in Fig. 2. If $a < b$, type-III order has easy-plane anisotropy and spins are in the xy plane. If $a > b$, the anisotropy is of easy-axis type and the spins are along the unique axis y. This is the case if the magnetic dipolar interaction is the dominating source of anisotropy.[52] More often, however, other sources of anisotropy dominate. Indeed, both easy-axis [K_2IrCl_6 (Refs. 48 and 49) and MnS_2 (Ref. 51)] and easy-plane [β-MnS (Ref. 50)] type-III structures have been found.

In order to understand the behavior of type-III antiferromagnets in an external field we study the transition at $B_c(T = 0)$ by using the soft-mode approach. If the anisotropy is of easy-plane type, the solution is simple. Antiferromagnetism develops at a vector belonging to the star of $\mathbf{Q} = \pi/a(1, 1/2, 0)$ when $B = B_c = S[\lambda(0) - \lambda_{1,3}(\mathbf{Q})]$ for all field alignments. In fields $0 < B < B_c$, the ground state is a canted four-sublattice structure in which the antiferromagnetic component is perpendicular to $\mathbf{B}$ and shows a repeated left-left-right-right pattern.[43]

The behavior of type-III antiferromagnets with easy-axis anisotropy is more complex. Because the easy axis is one of the three crystalline axes, this case corresponds, in fact, to the column for $\mathbf{Q} = (h, 0, 0)$ with $\lambda_{\min} = \lambda_1$. Of the high-symmetry-field directions, the transition takes place at a type-III structure for $\mathbf{B} \parallel [100]$ and $\mathbf{B} \parallel [110]$ when $B = B_c = S[\lambda(0) - \lambda_{1,3}(\mathbf{Q})]$. If the field is applied along the [111] crystalline direction, B_c will be lower and the ordering vector may or may not be of type III. We now concentrate on analyzing the soft-mode transition in this field direction.

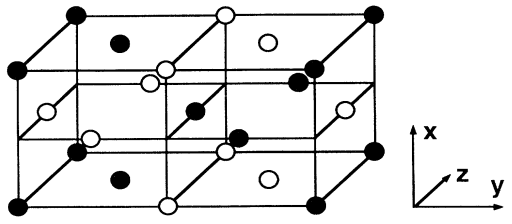

FIG. 2. Magnetic unit cell for type-III antiferromagnetic order in an fcc lattice. The black and white spheres represent oppositely directed spins for the ordering vector $\mathbf{k} = \pi/a(1, 1/2, 0)$. The spins are along the y axis in an easy-axis antiferromagnet, while they are along an arbitrary direction in the xz plane in an easy-plane system.

We assume that the Hamiltonian consists of an anisotropic nearest-neighbor (NN) interaction and an isotropic next-nearest-neighbor (NNN) interaction of the type $J_2\mathbf{S}_i \cdot \mathbf{S}_j$. The general form of the interaction between neighboring spins, separated by the lattice vector $\mathbf{r}_{ij} = (a, a, 0)$, is

$$\underline{A}[\mathbf{r}_{ij} = (a, a, 0)] = \begin{pmatrix} K_a & K_d & 0 \\ K_d & K_a & 0 \\ 0 & 0 & K_c \end{pmatrix} . \tag{26}$$

The components of $A_{ij}^{\alpha\beta}$ transform like $r_{ij}^{\alpha} r_{ij}^{\beta}$. For example, the interaction matrix for spins separated by $\mathbf{r}_{ij} = (a, 0, -a)$ reads

$$\underline{A}[\mathbf{r}_{ij} = (a, 0, -a)] = \begin{pmatrix} K_a & 0 & -K_d \\ 0 & K_c & 0 \\ -K_d & 0 & K_a \end{pmatrix} . \tag{27}$$

We next calculate the Fourier transform of the interaction matrix. It is necessary to find $\underline{A}(\mathbf{k})$ for wave vectors of the form $\mathbf{k} = (\pi/a, k_y, 0)$, which includes type-III ($k_y = \pi/2a$) and type-I ($k_y = 0$) ordering as particular cases. We find

$$\underline{A}(\pi/a, k_y, 0) = \begin{pmatrix} \lambda_1(k_y) & 0 & 0 \\ 0 & \lambda_2(k_y) & 0 \\ 0 & 0 & \lambda_3(k_y) \end{pmatrix} \tag{28}$$

with eigenvalues along the diagonal and eigenvectors $\hat{\mathbf{e}}_1 = (1, 0, 0)$, $\hat{\mathbf{e}}_2 = (0, 1, 0)$, and $\hat{\mathbf{e}}_3 = (0, 0, 1)$. The eigenvalues are

$$\begin{aligned} \lambda_1(k_y) &= -4K_a + 4(K_c - K_a)\cos(ak_y) \\ &\quad +2J_2[2 + \cos(2ak_y)] , \\ \lambda_2(k_y) &= -4K_c + 2J_2[2 + \cos(2ak_y)] , \\ \lambda_3(k_y) &= -4K_a - 4(K_c - K_a)\cos(ak_y) \\ &\quad +2J_2[2 + \cos(2ak_y)] . \end{aligned} \tag{29}$$

Note that K_d does not enter into the equations above. When $k_y = \pi/2a$ or $k_y = 0$ we recover the results by ter Haar and Lines.[3] We will not dwell on the question for which values of K_a, K_c, K_d, and J_2 the ground state is, in fact, a type-III structure. Here we just assume that the parameters are such that the zero-field structure is of type III.

In type-III magnets the $\cos(ak_y)$ term in Eqs. (29) vanishes. If $K_c > K_a$, the lowest eigenvalue is $\lambda_2(k_y)$ so that the structure has easy-axis anisotropy. When $J_2 = 0$, $\lambda_2(k_y)$ assumes the constant value $-4K_c$ along the line $\mathbf{k} = (\pi/a, k_y, 0)$. At $k_y = 0$, describing type-

I ordering, $\lambda_2 = \lambda_3$. The easy-axis type-III structure is hence degenerate with an easy-plane type-I state and with a continuum of easy-axis $\mathbf{k} = (\pi/a, k_y, 0)$ structures. A positive J_2 is needed to stabilize the type-III order.[3]

The degeneracy of the ground state for ordering vectors $\mathbf{k} = (\pi/a, k_y, 0)$ has been found previously for the NN antiferromagnetic Heisenberg model in the fcc lattice.[53] Our result shows that the degeneracy of the ordering vector is not lifted by the anisotropy of the NN interaction, although the degeneracy in the spin direction is. These results were found already by ter Haar and Lines[4] in the sense that they observed that there is no net average interaction between neighboring {100} planes, which are the building blocks of the $\mathbf{k} = (\pi/a, k_y, 0)$ spin structures.

A manifestation of the degeneracy—or near degeneracy—of the ordering vector is the suppression of the Néel temperature. Susceptibility measurements have indeed shown this to occur in K_2IrCl_6: The experimentally determined ratio of the Weiss temperature θ over the Néel temperature T_N is exceptionally large, $\theta/T_N = 10$.[54] The effect is well understood as the experimental θ/T_N ratio is in good agreement with the spin-wave calculation by Lines.[55]

The nearly degenerate energies of type-I and -III antiferromagnets for small J_2 are important in the selection of the spin structure in an external field. To find the soft mode for $\mathbf{B} \parallel [111]$, we first construct the 2×2 matrix $\underline{\tilde{A}}(\mathbf{k})$, which is the xy block of the 3×3 matrix $\underline{A}(\mathbf{k})$ with z denoting the direction of $\mathbf{B}$. Choosing $\hat{\mathbf{x}} = (1,-1,0)/\sqrt{2}$, $\hat{\mathbf{y}} = (1,1,-2)/\sqrt{6}$, and $\hat{\mathbf{z}} = (1,1,1)/\sqrt{3}$, we rotate $\underline{A}(\mathbf{k})$ into the basis $\{\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}}\}$ using Eq. (12). We find

$$\begin{aligned} A^{xx} &= (\lambda_1 + \lambda_2)/2, \\ A^{yy} &= (\lambda_1 + \lambda_2 + 4\lambda_3)/6, \\ A^{xy} &= (\lambda_1 - \lambda_2)/2\sqrt{3}\ . \end{aligned} \tag{30}$$

The eigenvalues $\tilde{\lambda}_{1,2}$ of $\underline{\tilde{A}}(\mathbf{k})$ are

$$\begin{aligned} \tilde{\lambda}_{1(2)}(k_y) = &-4[2K_a + K_c]/3 \mp 4[K_c - K_a] \\ &\times [1 + \cos^2(k_y)]^{1/2}/3 \\ &+2J_2[2 + \cos(2ak_y)]\ . \end{aligned} \tag{31}$$

For easy-axis anisotropy, $K_c - K_a > 0$ so that $\tilde{\lambda}_1 < \tilde{\lambda}_2$. Thus $\tilde{\lambda}_1$ is the relevant eigenvalue. For a type-III structure, $\tilde{\lambda}_1 = -4[K_a + 2K_c]/3 + 2J_2$, whereas $\tilde{\lambda}_1 = -4K_c + 6J_2$ for a type-I state. For $K_c - K_a > 3J_2$, spin waves with type-I wave vectors become soft at a higher field than those with type-III wave vectors. However, for the range of parameters $K_c - K_a \approx 3J_2$, soft modes with other $\mathbf{k} = (\pi/a, k_y, 0)$ vectors are possible as well. To find them we minimize $\tilde{\lambda}_1(k_y)$ along $(\pi/a, k_y, 0)$. The soft modes are

$$\begin{aligned} &K_c - K_a \le 2J_2: \quad \mathbf{q} = \pi/a(1, 1/2, 0)\ \text{(type III)}, \\ &2J_2 \le K_c - K_a \le 4J_2: \quad \mathbf{q} = [\pi/a, \cos^{-1}(\mu)/a, 0], \\ &K_c - K_a \ge 4J_2: \quad \mathbf{q} = \pi/a(1,0,0)\ \text{(type I)}, \end{aligned} \tag{32}$$

where

$$\mu = [(K_c - K_a)^2/12J_2^2 - 1/3]^{1/2}.$$

The antiferromagnetic structure is described by the above wave vector when $B \lesssim B_c$. For parameters $K_c - K_a > 2J_2$, a transition must take place in intermediate fields $0 < B < B_c$ because type-III order is stable at $B = 0$.

The first of Eqs. (32) provides an example of the inequality (21) in Sec. III B, describing the stability of the zero-field ordering vector. The strength of the anisotropic interactions has to be above a certain nonzero value for the ordering vector to change. Because of the constant eigenvalue along $\mathbf{k} = (\pi/a, k_y, 0)$ for NN interactions, the threshold in the present case does not depend on the isotropic NN interaction but only on J_2. As a consequence, the transition from an easy-axis type-III structure to a type-I state (perhaps via intermediate phases) in an external field can occur even when the spin-spin interactions are not pronouncedly anisotropic.

K_2IrCl_6 is a well-characterized antiferromagnet with $T_N = 3.05 \pm 0.03$ K.[56,54] Neutron-diffraction measurements have shown that the ordered structure at $B = 0$ is of type III, with spins along the unique cubic axis.[48,49] The nearest-neighbor Ir-Ir exchange interactions have been obtained from paramagnetic resonance[1,57] and susceptibility[54] measurements of a semidilute mixed crystal, where Pt was substituted for Ir. The measurements were analyzed in terms of a NN interaction

$$H_{\mathrm{NN}} = J\mathbf{S}_i \cdot \mathbf{S}_j + J_x S_i^x S_j^x + J_y S_i^y S_j^y + J_z S_i^z S_j^z \tag{33}$$

between NN Ir spins ($S = 1/2$). Here J is the isotropic part of the exchange interaction, while J_x, J_y, and J_z are the anisotropic terms, which satisfy $J_x + J_y + J_z = 0$. The subscripts x, y, and z denote the principal axes of the interaction. For nearest neighbors, separated by $\mathbf{r}_{ij} = (a, a, 0)$, x, y, and z are along (1,1,0), (1,$\bar{1}$,0), and (0,0,1), respectively.

For the exchange parameters it was found that $J/k_B = 11.5$ K, $J_x/k_B = -0.90$ K, $J_y/k_B = -0.39$ K, and $J_z/k_B = 1.29$ K.[1,57] The relations between these values and those in the interaction matrix (26) are

$$\begin{aligned} K_a &= J + (J_x + J_y)/2, \\ K_d &= (J_x - J_y)/2, \\ K_c &= J + J_z\ . \end{aligned} \tag{34}$$

The anisotropy $K_c - K_a$ is positive, as it should be for an easy-axis system.

The anisotropy of the NNN interaction was observed to be extremely small.[57] The isotropic part was found to be antiferromagnetic with $J_2/k_B = 0.55$ K. Thus type-III rather than type-I order is favored. The interactions are clearly NN dominated as $J_2/J_1 = 0.05$.

Assuming that the exchange parameters of the semidilute system and of the K_2IrCl_6 compound are the same, we have numerically determined the lowest eigenvalue of the Fourier transformed interaction matrix $\underline{A}(\mathbf{k})$ in a mesh of $12^3/2 = 864$ $\mathbf{k}$ vectors in the first Brillouin zone.

As expected, we find that $\lambda_{\min}$ is obtained at the type-III ordering vector $\mathbf{k} = (\pi/a, \pi/2a, 0)$. The measured interaction parameters give $(K_c - K_a)/J_2 = 3.5 \pm 0.5$. According to Eq. (32), we predict that for $\mathbf{B} \parallel [111]$, in a range of fields below B_c, the ordering vector is $\mathbf{k} = (\pi/a, k_y, 0)$ with $k_y \approx 0.2\pi/a$ although the order is of type III when $B = 0$. The error bars of $(K_c - K_a)/J_2$ are such that the predicted high-field k_y can vary from 0 to $0.28\pi/a$, thus also including type-I order.

Theoretical values for B_c are of experimental interest. When $\mathbf{B} \parallel [100]$ or $\mathbf{B} \parallel [110]$, $B_c = S^2[\lambda(0) - \lambda_{\min}]/g\mu_B$, where μ_B is the Bohr magneton and the factor $g = 1.79$.[1] We have here explicitly written the factor $\hbar\gamma = g\mu_B/S$, which was absorbed in B in the previous formulas. The eigenvalues are $\lambda(0) = 12J + 6J_2$ and $\lambda_{\min} = -4K_c + 2J_2$ so that $B_c = 40$ T. When $\mathbf{B} \parallel [111]$, B_c is lower by 1%. The predicted critical fields are somewhat high in view of the relatively low $T_N = 3.05$ K. This is because of the strong suppression of T_N in comparison with the Weiss temperature θ. The critical field is not expected to show a similar reduction as T_N.

Although the calculated B_c's are not yet readily reached in neutron experiments, one could perhaps study the stability of type-III order of K_2IrCl_6 when $\mathbf{B} \parallel [111]$ at temperatures close to T_N, where the critical field is lower. We expect, however, that the relative stability of the type-III structure increases along the $B_c(T)$ curve and the transition to another $\mathbf{k} = (\pi/a, k_y, 0)$ state might not take place at $T \lesssim T_N$, even though it occurred at $T = 0$. In any case, experiments probing the high-field spin structure of K_2IrCl_6 with $\mathbf{B} \parallel [111]$ seem interesting.

B. Nuclear ordering in copper

Susceptibility measurements have shown that nuclear spins in copper order antiferromagnetically below $T_N = 60$ nK.[25,58] Three antiferromagnetic phases were found as a function of the field below $B_c \approx 0.25$ mT.[59] The spin structures were later investigated by neutron diffraction. At zero field, the phases displayed type-I antiferromagnetic order, characterized by (1 0 0), (0 1 0), and (0 0 1) Bragg reflections.[60] As a function of the field, applied along the $[01\bar{1}]$ direction, the intensity of the (1 0 0) reflection first decreased and almost vanished at $B \approx 0.09$ mT. In still higher fields, the intensity of the (1 0 0) peak increased, showed a maximum around 0.16 mT, and finally vanished at $B_c = 0.25$ mT.[60] Another Bragg peak at (0 2/3 2/3) was found in intermediate and low fields, $0.01 < B < 0.13$ mT, with the maximum intensity around 0.10 mT.[26]

In more recent neutron-diffraction experiments, the magnetic field was applied also along the other crystallographic directions.[39] The general behavior of the various (1 0 0) and (0 2/3 2/3) reflections was the same as for $\mathbf{B} \parallel [01\bar{1}]$. In high fields, aligned close to the [100] crystalline directions, the (1 0 0) reflection was again found. However, for fields applied along the [111] direction, type-I order was not seen in the high-field region (Fig. 3), although the susceptibility signal indicated antiferromagnetism up to $B_c \approx 0.25$ mT. Neither did scans along several symmetric directions in the reciprocal space reveal any scattered neutrons. Experiments could not determine the ordered spin structure in this field region.

The spin-spin interactions in copper can be written as

$$A_{ij}^{\alpha\beta} = \frac{\mu_0}{4\pi}\hbar^2\gamma^2 r_{ij}^{-3}(\delta^{\alpha\beta} - 3r_{ij}^{\alpha}r_{ij}^{\beta}) + \delta^{\alpha\beta}J_{ij} + K_{ij}^{\alpha\beta} , \tag{35}$$

where the first term is the dipolar force, J_{ij} is the Ruderman-Kittel (RK) coupling, and $K_{ij}^{\alpha\beta}$ describes the anisotropic part of the exchange force. The RK term is isotropic in spin space. The interactions have been calculated from the electronic band structure. According to Lindgård, Wang, and Harmon[21] (LWH), assuming a nonrelativistic RK force, the minimum eigenvalue is at $\mathbf{k} = (\pi/a, 0, 0)$ and shows easy-plane anisotropy. Thus the high-field structure is of type I for all field directions in the soft-mode theory. According to the calculations by Oja, Wang, and Harmon[23] (OWH), which include both relativistic RK coupling as well as anisotropic terms, $\lambda_{\min}$ is at $\mathbf{k} = (0.87\pi/a, 0, 0)$, whereas $\lambda_{2,3}(\pi/a, 0, 0)$ is higher by 0.6%. Because $\lambda_{\min}$ is degenerate, the soft-

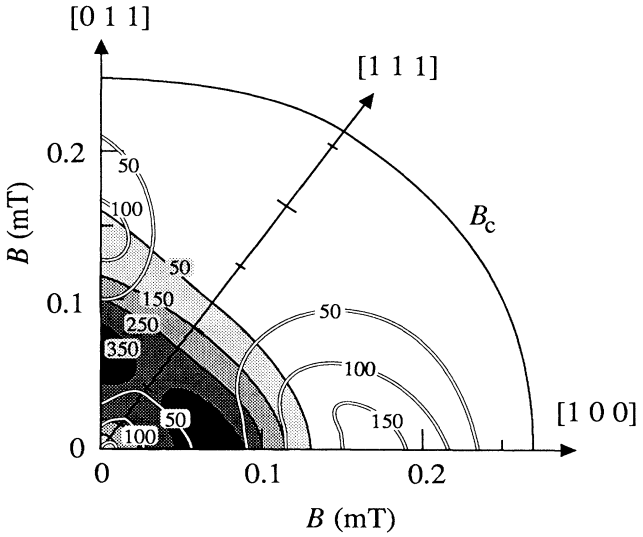

FIG. 3. Diagram illustrating the relative neutron intensities of the antiferromagnetic Bragg reflections (1 0 0) and (0 2/3 2/3) in copper as determined by measurements of Annila *et al.* (Ref. 39). The results are shown as a function of the magnetic field in the plane $B_y = B_z$, which contains the high-symmetry directions [011], [111], and [100]. The intensities for field directions equivalent under fcc symmetry have been summed together. The (1 0 0) intensity is marked by white contours and the region of the (0 2/3 2/3) intensity is shaded. The antiferromagnetic phase is bordered by the second-order B_c curve. In the [011] and [100] field directions, the (1 0 0) Bragg reflection was observed in fields immediately below $B = B_c = 0.27$ mT. For $\mathbf{B} \parallel [111]$, there is a large field region below $B = B_c$ in which no neutrons above the background were detected at the (1 0 0) and (0 2/3 2/3) Bragg positions. B_c for this field alignment was determined from the measurements of the low-frequency susceptibility.

mode transition always takes place at $\mathbf{k} = (0.87\pi/a, 0, 0)$, in disagreement with neutron-diffraction experiments.[39] The calculated spin-spin interactions, used within the soft-mode theory, cannot predict the unknown high-field phase.

Another problem with the theoretical exchange parameters is the stability of the (0 2/3 2/3) order: The (1 0 0) spin structure is favored in comparison to the (0 2/3 2/3) order in all magnetic fields. The balance between the ferromagnetic dipolar force and the antiferromagnetic RK interaction is, however, very delicate in copper as was shown by LWH.[21] Ordering vectors of the form $\mathbf{k} = (q, q, 0)$ with $q \approx 0.6$ have almost as low energy as the type-I order with $\mathbf{k} = (\pi/a, 0, 0)$. Thus the presence of (0 2/3 2/3) order in intermediate fields can be explained in two ways: (1) It is stabilized by fluctuations, in accordance with the theoretical prediction by Lindgård[61] or (2) by the molecular fields, which require that actually $\lambda(0, 2/3, 2/3) \lesssim \lambda(1, 0, 0)$, a situation that is within the estimated error limits of the band structure calculations.[21,23] The second possibility is the same as the basic assumption in our theoretical model,[40] which successfully explained the observed interplay of the (1 0 0) and (0 2/3 2/3) modulations[26] in copper within the mean-field theory and correctly predicted the selection rules for the various (0 2/3 2/3) reflections for different field alignments.[39] This encourages us to pursue the same approach in the present problem as well.

The eigenvalue $\lambda_n(\mathbf{k})$ has a double-well structure as a function of the $\mathbf{k}$-vector as was shown by LWH.[21] Although the band-structure calculations[21,23] indicate that $\lambda_{2,3}(1, 0, 0)$ is lower than $\lambda(0, 2/3, 2/3)$ by 10%, we assume here, as in our previous work,[40,41] that the modulation corresponding to the true $\lambda_{\min}$ is close to $\mathbf{k} = (\pi/a)(2/3, 2/3, 0)$ and that $\mathbf{k} = (\pi/a, 0, 0)$ is only a local minimum, i.e., $\lambda_{2,3}(1, 0, 0) \gtrsim \lambda(0, 2/3, 2/3) \gtrsim \lambda_{\min}$. An important principle in our previous analysis was to look for spin structures with only (1 0 0) and (0 2/3 2/3) modulations such that the lengths $|\mathbf{S}_i|$ of the moments are independent of the site i. For these structures, no higher harmonics with large eigenvalues λ are possible so that the energy is determined by the two small and almost equal eigenvalues $\lambda(0, 2/3, 2/3)$ and $\lambda_{2,3}(1, 0, 0)$. With (0 2/3 2/3) order only, this is possible at discrete values of the field such as $B = \sqrt{3/19}B_c$ when $B \parallel [111]$.[41] With a superposition of the (0 2/3 2/3) and (1 0 0) modulations, spins can have equal lengths in the range $0 \leq B \leq \sqrt{3/19}B_c$. It is the equal-moment principle that locks modulation exactly at (0 2/3 2/3)—there is no symmetry reason for $\lambda_{\min}$ to be located at this point. This is unlike the case for $\mathbf{k} = \pi/a(1, 0, 0)$ which, being a boundary point of the first Brillouin zone, can be the exact position of a local minimum for $\lambda_n(\mathbf{k})$.

At fields larger than $\sqrt{3/19}B_c$ when $B \parallel [111]$ it is not possible to combine (0 2/3 2/3) order to (1 0 0) to obtain equal-moment structures.[41] Therefore, the stabilizing lock-in mechanism for the (0 2/3 2/3) order does not exist. One can then expect that the high-field structure is of type I, as was concluded in earlier theoretical work,[61–65] except when the ordering vector corresponding to the true $\lambda_{\min}$ shows up for fields perpendicular to the eigenvector of $\lambda_{\min}$.

Let us next see what we can predict for the properties of the unknown phase, when $\mathbf{B} \parallel (1, 1, 1)$, on the basis of experimental results and the general properties of the soft-mode transition. We know that $\lambda_{2,3}(\pi/a, 0, 0) > \lambda_{\min}$ because otherwise the transition would always take place at a type-I vector. In addition, $\lambda_{\min}$ must be a unique eigenvalue; otherwise the mode would become soft for all directions of $\mathbf{B}$. The corresponding eigenvector $\mathbf{e}$ should not be perpendicular to any of the three [100] directions. Therefore, $e_x \neq 0$, $e_y \neq 0$, $e_z \neq 0$. Neither should $\mathbf{e}$ be perpendicular to any of the six [110] directions. Thus, $|\, e_x \,| \neq |\, e_y \,|$, $|\, e_x \,| \neq |\, e_z \,|$, and $|\, e_y \,| \neq |\, e_z \,|$.

Inspecting Table I, we then find that the only possible ordering vector satisfying these conditions is of the most general form, viz., $\mathbf{Q} = (h, k, l)$, with $|h|$, $|k|$, and $|l|$ all unequal and nonzero. To stabilize antiferromagnetic order with a $\mathbf{Q} = (h, k, l)$ when $\mathbf{B} \parallel (1, 1, 1)$, the eigenvector $\mathbf{e}$, corresponding to $\lambda_{\min}$, has to be almost perpendicular to (1,1,1). These conclusions are identical to our previous predictions based on the mean-field theory.[43]

We have confirmed that the $\mathbf{Q} = (h, k, l)$ phase for $\mathbf{B} \parallel [111]$ is not unreasonable in view of the band-structure calculations of the exchange interactions. By slightly varying the interaction parameters from their calculated values, we have produced a set of parameters for which $\lambda_{\min}$ is obtained at the general, $\mathbf{Q} = (h, k, l)$ type of position in the $\mathbf{k}$ space. Table II shows these parameters, together with the original figures calculated by OWH.[23] The largest absolute change was made in the nearest-neighbor interaction which has been enhanced by 18%.

We shall next show that the modified Hamiltonian gives the experimentally observed high-field behavior, calculated within the soft-mode theory. Because we intend to employ the Hamiltonian in a numerical simulation as well,[66] we have included the dipolar interaction up to the eighth-nearest neighbors only and accounted for the longer ranging forces by the demagnetization and Lorenz factors. For the same reason, the set of $\mathbf{k}$ vectors has been restricted to those compatible with a system of $12^3/2 = 864$ spins in an fcc lattice. Within this set, $\lambda_{\min}/k_B = -135.57$ nK is obtained at $\mathbf{k} = (\pi/a)(2/3, 1/2, 1/6)$. The eigenvalues for the other two relevant $\mathbf{k}$ vectors are $\lambda[(\pi/a)(1, 0, 0)]/k_B = -135.16$ nK and $\lambda[(\pi/a)(0, 2/3, 2/3)]/k_B = -135.31$ nK. For any other $\mathbf{k}$, $\lambda_n(\mathbf{k})$ has a value higher than these. The eigenvalue for $\mathbf{k} = 0$, which is needed to calculate B_c, is $\lambda(0)/k_B = 0.87$ nK, assuming zero for the demagnetization factor. The eigenvalues for the (1 0 0) and (0 2/3 2/3) order are such that their interplay in low fields produces consistency with experiments[26] and our earlier calculations.[40] Our results depend on the particular choices for the interaction parameters only through the three eigenvalues $\lambda[(\pi/a)(1, 0, 0)]$, $\lambda[(\pi/a)(0, 2/3, 2/3)]$, and $\lambda[(\pi/a)(2/3, 1/2, 1/6)]$, and the direction of the eigenvector $\mathbf{e}\,[(\pi/a)(2/3, 1/2, 1/6)]$.

The critical field $B_c(2/3, 1/2, 1/6) = S[(\lambda(0) -$

TABLE II. Elements of the total exchange interaction matrix $(A_{ij}^{\rm ex})^{\alpha\beta} = \delta^{\alpha\beta}J_{ij} + K_{ij}^{\alpha\beta}$, Eq. (35), for different lattice vectors $\mathbf{r}_{ij}$. The values in the first columns are first-principles calculations by OWH (Ref. 23) second columns are the modified interactions. The values are given in nK. The parameters $R = \sum_j A_{ij}^{\rm ex}/(\mu_0\hbar^2\gamma^2\rho)$ and $Q = [\sum_j (A_{ij}^{\rm ex})^2]^{1/2}/(\mu_0\hbar^2\gamma^2\rho)$, where $A_{ij}^{\rm ex} = \frac{1}{3}[(A_{ij}^{\rm ex})^{xx} + (A_{ij}^{\rm ex})^{yy} + (A_{ij}^{\rm ex})^{zz}]$ and ρ is the number density of atoms, are (R, Q) = (i) (0.34, 0.092) as calculated by OWH;[23] (ii) (0.34, 0.105) for the modified interactions; (iii) (0.42 ± 0.05, 0.095 ± 0.003) as determined by NMR measurements (Refs. 17 and 16.)

$\mathbf{r}_{ij}$	$(A_{ij}^{\rm ex})^{xx}$		$(A_{ij}^{\rm ex})^{yy}$		$(A_{ij}^{\rm ex})^{zz}$		$(A_{ij}^{\rm ex})^{xy}$		$(A_{ij}^{\rm ex})^{xz}$		$(A_{ij}^{\rm ex})^{yz}$	
	OWH	Modif.	OWH	Modif.	OWH	Modif.	OWH	Modif.	OWH	Modif.	OWH	Modif.
(1,1,0)	12.597	14.884	12.597	14.884	9.428	10.958	1.979	0.644	0	0	0	0
(2,0,0)	-1.649	-1.337	-1.802	-1.507	-1.802	-1.507	0	0	0	0	0	0
(2,1,1)	2.003	0.637	2.005	0.664	2.005	0.664	-0.172	-0.197	-0.172	-0.197	0.249	0.130
(2,2,0)	0.320	0.530	0.320	0.530	0.272	0.495	-0.442	-0.234	0	0	0	0
(3,1,0)	-0.580	-0.580	-0.487	-0.487	-0.714	-0.714	0.008	0.008	0	0	0	0
(2,2,2)	-0.945	-1.085	-0.945	-1.085	-0.945	-1.085	-0.034	-0.034	-0.034	-0.034	-0.034	-0.034
(3,2,1)	-0.120	-0.087	-0.098	-0.065	-0.117	-0.084	0.046	0.046	-0.026	-0.026	0.050	0.050
(4,0,0)	0.350	0.350	0.350	0.350	0.350	0.350	0	0	0	0	0	0
(4,1,1)	0.030	0.000	0.030	0.000	0.030	0.000	0	0	0	0	0	0

$\tilde{\lambda}(2/3, 1/2, 1/6)]$ reaches its maximum when $\tilde{\lambda}(h,k,l) = \lambda_{\rm min}$, which occurs when $\mathbf{B}$ is perpendicular to any of the vectors in the star of the corresponding eigenvectors $\{\hat{e}\}^* = \langle e_x, e_y, e_z\rangle$. Thus there are 24 planes $\{e_x, e_y, e_z\}$, shown in Fig. 4(a), such that $B_c(2/3, 1/2, 1/6)$ is maximized for fields in these planes. The angle between the eigenvector $\mathbf{e} = (-0.5369, 0.8145, -0.2199)$ and the [111] direction is 88.09°, so that they are almost perpendicular. There are six $\{e_x, e_y, e_z\}$ planes close to the [111] direction, whereas there are none in the vicinity of the [001] and [011] directions. Thus $B_c(2/3, 1/2, 1/6)$ has a low value for $\mathbf{B} \parallel$ [001] and $\mathbf{B} \parallel$ [011] but a high value for $\mathbf{B} \parallel$ [111].

To see how the critical fields for the ordering vectors in the star of $\mathbf{k} = (\pi/a)(2/3, 1/2, 1/6)$ vary between the maximum values we calculated $B_c(2/3, 1/2, 1/6) = S[(\lambda(0) - \tilde{\lambda}(2/3, 1/2, 1/6)]$ as a function of the field direction. It is then necessary to know also the two larger eigenvalues and their eigenvectors for the Fourier transformed interaction matrix $\underline{A}[\mathbf{k} = (\pi/a)(2/3, 1/2, 1/6)]$. These can be obtained from

$$\underline{A}(2/3, 1/2, 1/6) = \begin{pmatrix} 5.69 & 107.51 & 53.28 \\ 107.51 & -47.23 & 64.69 \\ 53.28 & 64.69 & -26.09 \end{pmatrix}, \qquad (36)$$

which is given in units of $\mathrm{nK}k_B$. Figure 5 shows the calculated $B_c(2/3, 1/2, 1/6)$ for fields in the plane $B_y = B_z$, which contains the high-symmetry directions [100], [111], and [011]. There are six "fingers" (A)–(F) centered at the directions in which the different $\{e_x, e_y, e_z\}$ planes intersect the plane $B_y = B_z$. The widths of the fingers depend on the angles of intersection. Note that the resolution has been enhanced greatly to display the dependence of B_c on the field direction.

The critical fields for the $\mathbf{k} = (\pi/a)(1, 0, 0)$ and $\mathbf{k} = (\pi/a)(0, 2/3, 2/3)$ ordering vectors have been included in Fig. 5. Both $B_c(1, 0, 0)$ and $B_c(0, 2/3, 2/3)$ are constant in the plane $B_y = B_z$, the former owing to the degeneracy of $\lambda(1, 0, 0)$ and the latter because the corresponding eigenvector $(1/\sqrt{2})(0, 1, -1)$ is perpendicular to this plane [see Fig. 4(b)]. All other modulations in the mesh of 864 $\mathbf{k}$ vectors yield lower critical fields. According to the diagram, the soft-mode transition around the [111] field direction takes place at an antiferromagnetic state described by the ordering vector $\mathbf{k} = (\pi/a)(2/3, 1/2, 1/6)$ or its cubic equivalents. This explains why neither (1 0 0) nor (0 2/3 2/3) Bragg reflection was observed in high fields when $\mathbf{B} \parallel$ [111]. Around the [100] and [011] field alignments, the (0 2/3 2/3) modulation should appear. $B_c(0, 2/3, 2/3)$ is, however, sensitive to any misalignment of the field, whereas $B_c(1, 0, 0)$ is independent of the field direction. If $\mathbf{B}$ is tilted by 2° off the plane $B_y = B_z$, the (1 0 0) order is stabilized over (0 2/3 2/3). Moreover, when the field is decreased below B_c, the equal-moment principle favors (1 0 0) order against (0 2/3 2/3). It is, therefore, understandable that in the experiments it is the (1 0 0) order instead of (0 2/3 2/3), which is seen in high fields close to the [1 0 0] and [0 1 1] directions.

In the above example, $\lambda_{\rm min}$ was obtained at $\mathbf{k} = (\pi/a)(2/3, 1/2, 1/6)$, but the true position $\mathbf{k} = (h, k, l)$ is unknown. We now state briefly the general properties of a finger diagram like that of Fig. 5 for any modulation

of the (h,k,l) type. Let us denote by θ the angle of the field with respect the [011] direction: $\theta = \theta_{[111]} = 35.26°$ for a field in the [111] direction and $\theta = 90°$ for a field along [100]. For vectors in the star of the eigenvector $\mathbf{e} = (\alpha,\beta,\gamma)$, $0 < \alpha < \beta < \gamma$, the positions of the six maxima as functions of the eigenvector components are

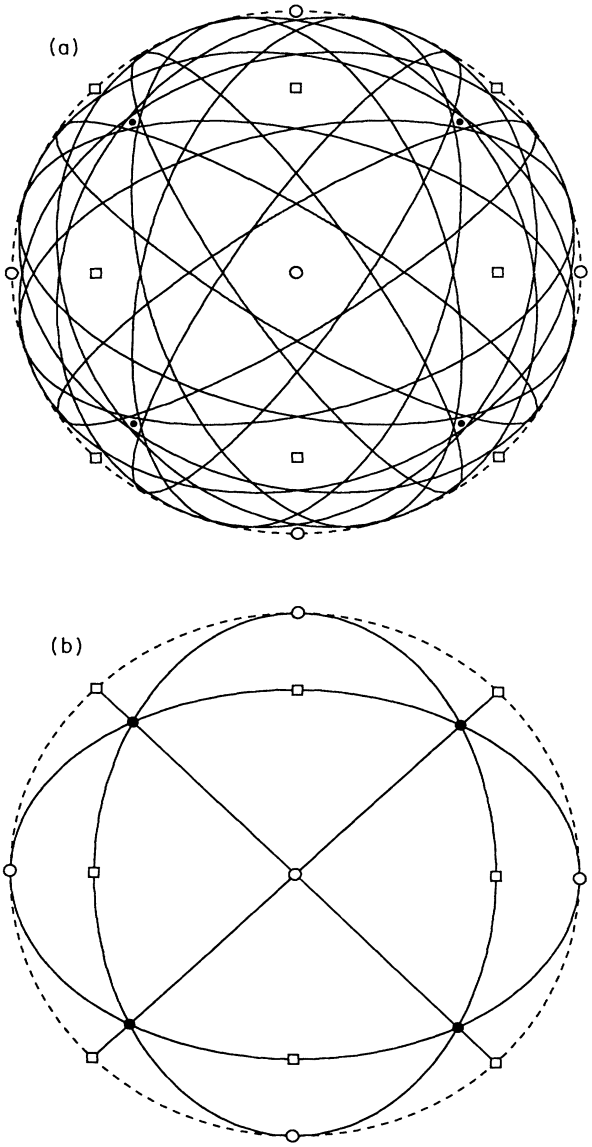

FIG. 4. Directions of the magnetic field $\mathbf{B} = (B_x, B_y, B_z)$ for which the critical field B_c of the antiferromagnetic modulation reaches its maximum value according to our calculations. Full lines are formed by the corresponding points $(B_x/B, B_y/B, B_z/B)$ on the unit sphere $x^2 + y^2 + z^2 = 1$, projected onto the xy plane. The symbols (○), (□), and (•) mark the high-symmetry crystalline directions [100], [110], and [111], respectively. (a) Modulations described by the ordering vectors in the star of $\mathbf{Q} = \pi/a(2/3, 1/2, 1/6)$ with spin direction $\hat{\mathbf{e}}(\mathbf{Q}) = (-0.5369, 0.8145, -0.2199)$; (b) the same for $\mathbf{Q} = \pi/a(0, 2/3, 2/3)$ with $\hat{\mathbf{e}}(\mathbf{Q}) = \frac{1}{\sqrt{2}}(0, 1, -1)$.

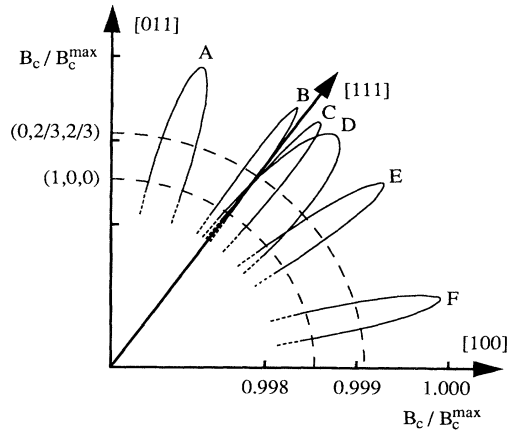

FIG. 5. Critical fields B_c for antiferromagnetic structures with ordering vectors in the star of $\mathbf{Q} = \pi/a(2/3, 1/2, 1/6)$ as a function of the external field direction in the plane $B_y = B_z$. The scale is $B_c^{max} = 0.37$ mT. The finger-shaped curves labeled A,. . ., F represent critical fields for the various ordering vectors $\mathbf{Q} = (h,k,l)$ as expressed by Eq. (38), with $h = 2\pi/3a$, $k = \pi/2a$, and $l = \pi/6a$. The resolution has been greatly enhanced as shown by the scale on the horizontal axis. The broken lines with labels $(1,0,0)$ and $(0,2/3,2/3)$ show B_c's for the corresponding ordering vectors.

$$\cos(\theta) = \frac{|e_x|}{\sqrt{\frac{1}{2}(e_y + e_z)^2 + e_x^2}}\ , \tag{37}$$

where

$$\begin{aligned}
&(A)\ \hat{\mathbf{e}} = (\gamma,\alpha,-\beta),\quad \mathbf{Q} = (l,h,-k),\\
&(B)\ \hat{\mathbf{e}} = (\gamma,-\alpha,-\beta),\quad \mathbf{Q} = (l,-h,-k),\\
&(C)\ \hat{\mathbf{e}} = (\beta,-\gamma,\alpha),\quad \mathbf{Q} = (k,-l,h),\\
&(D)\ \hat{\mathbf{e}} = (\alpha,\beta,-\gamma),\quad \mathbf{Q} = (h,k,-l),\\
&(E)\ \hat{\mathbf{e}} = (\beta,-\gamma,-\alpha),\quad \mathbf{Q} = (k,-l,-h),\\
&(F)\ \hat{\mathbf{e}} = (\alpha,-\beta,-\gamma),\quad \mathbf{Q} = (h,-k,-l)\ .
\end{aligned} \tag{38}$$

The above vectors $\hat{\mathbf{e}}$ can be reflected in the plane $B_y = B_z$ and/or inverted, while retaining the value of θ. Therefore, each maximum of B_c corresponds to four different reflections of the (h, k, l) type, amounting to a total of 24 reflections. The other 24 (h, k, l) reflections correspond to symmetrically equivalent values of $\theta \in [\pi/2, \pi]$. The relative positions of the six fingers satisfy the inequalities

$$\begin{aligned}
&\theta_A < \theta_B < \theta_E < \theta_F\ ,\\
&\theta_C < \theta_E,\\
&\theta_D < \theta_F\ .
\end{aligned} \tag{39}$$

If the stability criterium of Eq. (31) is not satisfied at $\mathbf{Q} = (h,k,l)$, the ordering vector may move in the $\mathbf{Q}$ space when the field is turned, and a more complicated phase diagram results.

Several remarks should be made when the theoretical diagram of Fig. 5 is compared with its experimental

counterpart, Fig. 3. The soft-mode theory applies only at the phase boundary. It is not known how far into the antiferromagnetically ordered region the stability regions of the (h,k,l) phases extend to. One expects that the more the boundary of a finger bulges out, the deeper inside the antiferromagnetic region the (h,k,l) state is stable. Below B_c, the characteristics of the particular spin configuration, in addition to the value of $\lambda(h,k,l)$, are important as well: The configuration can be a single-$\mathbf{k}$ or a multiple-$\mathbf{k}$ structure, and higher harmonics may be induced. In the plane $B_y = B_z$, the most stable (h,k,l) structure is a $4-\mathbf{k}$ state although for $\mathbf{B} \parallel [111]$ a $6-\mathbf{k}$ state is also possible.[43,66,67] These questions will be investigated by numerical simulations in a forthcoming paper.[66]

The theoretical phase diagram suggests that in high fields aligned close to the [111] direction there are in fact several phases. This explains the large field region with no observed neutron intensity at the (100) Bragg position. When the diagrams are compared on a finer scale, it seems as if the experimental diagram would not support a (h,k,l) state corresponding to the (F) finger. There is no real discrepancy, however, because the neutron measurements in field directions between the three high-symmetry directions [100], [111], and [011] were conducted only in fields $B = 0.09$ and 0.16 mT, which are clearly below $B = B_c = 0.25$ mT. The neutron contours just interpolate these data and more extensive results in the high-symmetry directions. Note also that experimental results for $\mathbf{B}$ aligned in between the directions [100], [111], and [011] may suffer from effects due to metastability and slow nucleation of the thermodynamically stable state,[68] whereas the data for the three symmetry directions are less likely to suffer from nonequilibrium effects as these directions were investigated in more extensive measurements.[69] Because of the agreement between our theory and the experiments for $\mathbf{B} \parallel [100]$, $\mathbf{B} \parallel [111]$, and $\mathbf{B} \parallel [011]$, we conclude that neutron-diffraction data support our calculation and that it is worthwhile experimentally to look for the predicted fine structure in the phase diagram.

The high-field spin structure of copper for $\mathbf{B} \parallel [111]$ has recently been studied also by Lindgård.[42] Although he too employed the soft-mode theory, a detailed comparison between his and our results is not possible because Lindgård has reported only his final numerical results. Lindgård found that the wave vector of the spin-wave excitation in the paramagnetic state, which softens is (i) the type-I vector $[\pi/a(1,0,0), \pi/a(0,1,0)$, or $\pi/a(0,0,1)]$ when $\mathbf{B} \parallel [001]$ or $\mathbf{B} \parallel [110]$, and (ii) $\mathbf{k} = (t+\delta, t-\delta, 0)$ with $t \sim (2/3)(\pi/a)$ and $\delta \sim 0.05(\pi/a)$ when $\mathbf{B} \parallel [111]$. These results are, however, in contradiction with the analytical properties of the soft modes (Sec. III). It is seen from Table I, that if the ordering vector $\mathbf{Q}$ is of the form $(h,k,0)$, there is a soft-mode transition at this wave vector or its cubic equivalent when $\mathbf{B} \parallel [100]$. This is also true if $\mathbf{Q}$ is a type I vector. To obtain a soft-mode transition at a type-I rather than a type-$(h,k,0)$ vector for $\mathbf{B} \parallel [100]$, the eigenvalues must be such that $\lambda(\pi/a,0,0) < \lambda(h,k,0)$. In copper, the type-I order has easy-plane anisotropy: $\lambda_2(\pi/a,0,0) = \lambda_3(\pi/a,0,0)$. Because of this degeneracy, the transition will then occur for a type-I vector in an arbitrary field direction, in contradiction with the finding (ii). Our numerical results confirm that Lindgård's calculations must suffer from an error.[70]

C. Other systems

Nuclear ordering has recently been investigated in silver as well.[18,19,27–29,71,72] Antiferromagnetic order was observed below 560 pK.[27] Experiments have been extended to negative spin temperatures as well, where ferromagnetic order has been found at $T > -1.9$ nK.[72] The spin-spin interactions of silver have been studied extensively both experimentally[9,18,19,71,73] and theoretically.[9,24,74] The RK interaction is antiferromagnetic and clearly dominates over the dipolar force. The anisotropic exchange interactions should be small in comparison to the dipolar term.[74] The spin-spin Hamiltonian for silver is thus rather isotropic with the dipolar interaction being the largest anisotropic term.

The predicted ground state of silver at zero field is a type-I antiferromagnet, characterized by the ordering vectors $(\pi/a,0,0)$, $(0,\pi/a,0)$, and $(0,0,\pi/a)$ and with easy-plane anisotropy.[24,74,75] The soft-mode analysis then predicts, consistently with earlier calculations using the mean-field[62] and spin-wave[63] theories and Monte Carlo simulations,[64,76] that the transition from the polarized paramagnetic state takes place to a type-I antiferromagnet for all alignments of the external field. These predictions can, hopefully, soon be tested in planned neutron-diffraction experiments.[77]

Studies of nuclear ordering in gold are underway.[36] In this metal, the RK interaction is expected to be even stronger than in silver.[63] The predicted spin structure is again of type I. Although weak in comparison with the isotropic RK interaction, the strength of the anisotropic exchange might be comparable to dipolar forces. It cannot, therefore, be predicted whether the expected type-I structure shows easy-plane anisotropy as in copper and silver or easy-axis anisotropy as in some lanthanide and actinide monopnictides with type-I order.[2] The magnetic phase diagram would be more interesting for the easy-axis case because, for example, with the [111] alignment of the magnetic field, there would be competition between the energies caused by anisotropy and external field.

Among the other metallic nuclear magnets investigated so far,[30–38] the soft-mode theory has recently been used by Aoyama and Ishii[78] to investigate field-induced phase transitions in rare-earth compounds such as $PrNi_5$. An extension of the present analysis to the hexagonal symmetry would allow studies of Pr, Tl, and Sc as well. Although $AuIn_2$ has a cubic structure, we cannot draw any conclusions about nuclear ordering in this compound because the spin-spin interactions are not known. Rh provides an interesting material for a study of magnetic order in an external field, since the interactions are strongly anisotropic.[74] Reliable prediction of the ordered structure calls for first-principles calculation of the exchange interactions.

VI. SUMMARY AND DISCUSSION

The transition from the polarized paramagnetic state to the antiferromagnetic phase in the critical field B_c at $T = 0$ has been investigated by spin-wave theory. The analysis applies to spins in a lattice of cubic symmetry. The Hamiltonian consists of the Zeeman term and spin-spin interactions, which can be anisotropic. By employing the linear spin-wave theory, antiferromagnetic transition to a state described by an ordering vector $\mathbf{Q}$ was discussed in terms of softening of the corresponding spin-wave excitation.

The onset of antiferromagnetic order for $B = B_c$ was shown to be an eigenvalue problem, which takes the same form both in the soft-mode and mean-field theories. The smallest eigenvalue of the Fourier transformed 2×2 interaction matrix, which describes the spin-spin interactions in the plane $\perp \mathbf{B}$, determines B_c, $\mathbf{Q}$, and the spin direction. For a sufficiently anisotropic spin-spin interaction, all these quantities can depend on the direction of $\mathbf{B}$ with respect to the crystalline axes. However, when $\mathbf{Q}$ shows an easy-plane anisotropy, which is possible for ordering vectors of the type $\mathbf{Q} = (h, 0, 0)$ and $\mathbf{Q} = (h, h, h)$, the direction of $\mathbf{B}$ has no such effect. The number of modes that become simultaneously soft when $B = B_c$ was tabulated for the various symmetry directions of $\mathbf{Q}$ and $\mathbf{B}$ which are possible in a cubic system.

The general results were first applied to study the stability of the easy-axis type-III antiferromagnetism of an fcc lattice. It was shown that when the anisotropic nearest-neighbor interaction is much stronger than the isotropic next-nearest-neighbor coupling, type-III order is unstable against type-I order in a sufficiently high magnetic field applied along the [111] crystalline axis. When the anisotropic and isotropic terms are comparable, the high-field ordering vector is of the type $\mathbf{Q} = (\pi/a, k_y, 0)$, which is between type-I and type-III vectors. Such an ordering vector, with $k_y \approx 0.2\pi/a$, was predicted for K_2IrCl_6 in the high-field region $B \lesssim B_c$ when $\mathbf{B} \parallel [111]$. Experiments were proposed to look for this transition.

Another application discussed in this paper was the magnetic phase diagram of nuclear spins in copper at nanokelvin temperatures. We looked for an explanation for copper not showing type-I antiferromagnetism in the region of fields below B_c when $\mathbf{B} \parallel [111]$, although this type of ordering was observed in the same fields when $\mathbf{B} \parallel [100]$ or [110]. The only explanation compatible with the soft-mode theory is that for $\mathbf{B} \parallel [111]$ softening of the spin-wave mode takes place at a wave vector $\mathbf{Q} = (h, k, l)$, where $|h|$, $|k|$, and $|l|$ are all unequal and nonzero. The result agrees with our previous proposal based on the mean-field theory.[43] It was shown that various (h, k, l) phases should be stable in the high-field region $B \lesssim B_c$ for several other field alignments besides those close to [111]. A model Hamiltonian reproducing the experimentally observed behavior at high fields was constructed with exchange parameters close to their theoretically calculated values. In a forthcoming paper,[66] we will describe the spin configurations in the whole ordered region, determined from numerical simulations using the same Hamiltonian. Some remarks were made on the ordered nuclear spin structures of silver and gold, which are expected to be of type I.

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[68]In Fig. 3, in regions between the three high-symmetry directions, the neutron contours may be affected by metastability and history dependence of the spin system, as shown by the data at $B = 0.16$ mT in Fig. 18 of Ref. 39.
[69]Experimental data in Figs. 13–16 of Ref. 39.
[70]Using the same model Hamiltonian as Lindgård in Ref. 42, namely $J_1/k_B = -16.65$ nK, $J_2 = -J_1/10$, neglecting J_{ij}'s for more distant neighbors, and including dipolar interactions up to infinity (with $\gamma^2\hbar^2 r_{ij}^{-3}/k_B = 25.43$ nK corresponding to $R = -0.42$) we obtain the minimum eigenvalue $\lambda_{\min}/k_B = -161$ nK with the eigenvector (1, −1, 0) at $\mathbf{k} = \pi/a(3/4, 3/4, 0)$. This agrees with Lindgård's unpublished results (private communication). Our numerical calculations confirm that the soft-mode transition takes place at this **k** vector when $\mathbf{B} \parallel [001]$, $\mathbf{B} \parallel [110]$, and $\mathbf{B} \parallel [111]$, in agreement with the analytical results of Sec. III A but in disagreement with Lindgård's numerical results.
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