Spin-wave dispersion and exchange interactions in $Y_{0.90}Tb_{0.10}$ and $Y_{0.90}Ho_{0.10}$ alloys*

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Neutron inelastic scattering experiments carried out on $Y_{0.90}Tb_{0.10}$ and $Y_{0.90}Ho_{0.10}$ at 4.7 °K show that well-defined spin waves exist in dilute alloys of the heavy rare earths Tb and Ho in yttrium metal. The measured spin-wave dispersion relations are satisfactorily described by a mean-lattice model together with the assumption that the exchange interaction $\mathcal{J}(q)$ is related to the conduction-electron susceptibility $\chi(\vec{q})$ of yttrium and, hence, is common to both alloys. The $\mathcal{J}(\vec{q})$ function deduced from the data possesses a peak, at the wave vector of the helical magnetic structures of the alloys, that is much larger than those measured for the pure rare-earth metals. This result is consistent with theoretical calculations of $\chi(\vec{q})$ by Liu, Gupta, and Sinha from the electronic energy bands for yttrium and with measurements of the magnetic structures of yttrium–rare-earth alloys as a function of composition and applied magnetic field.

I. INTRODUCTION

The heavy-rare-earth metals are known to possess very complicated magnetic structures which are determined primarily by an indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) type of exchange interaction. The magnetic moment carried by highly localized $4f$ electrons on each ion interacts with other moments by polarizing the spins of the conduction electrons surrounding the ions. Thus, the RKKY interaction is governed by the exchange interaction U_q between the localized spin moment and a conduction-electron spin and by the response of the conduction electrons to the effective field of the magnetic moment. This response is given by the generalized susceptibility $\chi(\vec{q})$, which is determined by the conduction-electron energy bands of the metal. In terms of U_q and $\chi(\vec{q})$ the Fourier transform of the indirect exchange interaction $j(\vec{q})$ between rare-earth ions is given approximately by[1]

$$j(\vec{q}) = U_q^2 \chi(\vec{q}) . \tag{1}$$

Therefore, information about $\chi(\vec{q})$ may be obtained from $j(\vec{q})$, which can be determined experimentally from measurements of the spin-wave spectra for various rare-earth metals.

Yttrium, which forms continuous solid solutions with all the heavy rare earths, has an outer electron configuration similar to the rare earths but possesses no magnetic moment. It is therefore considered to be an ideal nonmagnetic solvent which merely increases the average separation of rare-earth ions while leaving the RKKY interaction unchanged. However, a few experimental observations seem to indicate that the RKKY interaction does change appreciably by dilution with yttrium.[2,3] In contrast to the variety of magnetic structures of the pure metals, alloys of yttrium with low concentrations of heavy rare earths always exhibit

oscillatory antiferromagnetic structures and ordering temperatures which decrease by dilution with yttrium.[3] For example, at low temperatures Gd, Tb, Ho, and Dy alloys all exhibit a spiral structure with the turn angle of the magnetic moment of 50° in the low-concentration limit. Thus the generalized susceptibilities for such alloys are probably very similar, and may be regarded to be almost the same as that of pure yttrium.

As in the case of pure rare-earth metals, information about the exchange interaction may be obtained from neutron inelastic scattering measurements of the spin-wave dispersion relations for these alloys. Since the RKKY interaction is long range, it is not possible to derive an accurate expression for the spin-wave dispersion relations of these random magnetic alloys, in contrast to the case of magnetic insulators.[4] However, the very fact that the interaction is long range may make the effects of variations of the local environment around different magnetic ions less important and justify the use of a mean-lattice approximation in which each atomic site is assumed to be occupied by an average atom as discussed in the Appendix. In this approximation, the equations for spin waves can be obtained by averaging the equations of motion for spins in the lattice over all the atomic sites. The resulting equations recover the symmetries of the hcp structure and lead to the expression for spin-wave energies in the c direction given by

$$\hbar\omega(q) = J([\, c\,\mathcal{J}(\vec{q}_0) - c\,\mathcal{J}(\vec{q}) + 2B]\{ c\,\mathcal{J}(\vec{q}_0)$$

$$- \tfrac{1}{2}[c\,\mathcal{J}(\vec{q}_0 + \vec{q}) + c\,\mathcal{J}(\vec{q}_0 - \vec{q})]\})^{1/2}, \tag{2}$$

where J is the total angular momentum, c is the concentration of magnetic ions, B is the crystal-field anisotropy constant, and $\vec{q}_0$ is the wave vector which characterizes the spiral structure. As

one might expect, this result is identical to the
expression for spin-wave energies of the rare-
earth metals having spiral magnetic structures,
but with the exchange function $\mathcal{J}(\vec{q})$ reduced by the
factor c. Furthermore, the strong spin-orbit
coupling in the $4f$ shell of the rare earths has the
effect that the spin $\vec{S}$ is not a constant of the motion.
Thus, as usual, the Hamiltonian from which Eq.
(2) is derived is obtained by projecting $\vec{S}$ on $\vec{J}$,
i.e., $\vec{S} = (g - 1)\vec{J}$, so that the exchange $\mathcal{J}(\vec{q})$
$= (g - 1)^2 j(\vec{q})$, where g is the Landé factor.

II. EXPERIMENT

A triple-axis neutron spectrometer at the Oak
Ridge High Flux Isotope Reactor was used to mea-
sure the spin-wave dispersion curves at 4.7 °K for
two yttrium–rare-earth alloys, one containing 9.8-
at.% Tb and the other containing 9.5-at.% Ho.
The single-crystal samples were grown by the
strain-anneal method. The Néel temperatures
were observed to be 54 °K for Y(Tb) and 20 °K for
Y(Ho). The (101) planes of Be crystals were used
as monochromator and analyzer in all the measure-
ments, and Soller slits with 40' angular resolution
were placed before and after the sample. The
measurements were performed with the energy of
the scattered neutrons fixed at 12.8 meV, and pyro-
lytic graphite filters were placed between the sam-
ple and the analyzer. Scans were carried out,
with either the neutron momentum change $\hbar\vec{Q}$ or the
neutron energy change $\hbar\omega$ held constant, for $\vec{Q}$ near
the 002 reciprocal lattice point of the sample and
for $\vec{q}$ along the [001] direction.

The results of some of the scans for Y(Ho) are
shown in Fig. 1. For the spiral magnetic struc-
tures studied in this work, two spin-wave disper-
sion curves, with their origins at $\vec{\tau} \pm \vec{q}_0$ for each re-
ciprocal lattice vector $\vec{\tau}$, are visible in neutron
scattering experiments when $\vec{Q}$ is in the [001] direc-
tion.[5] Thus two peaks are expected in the distri-
bution of scattered neutrons, as illustrated in Fig.
1(b), except at points where the curves cross, e.g.,
$(0, 0, 3)2\pi/c$, or at points where one curve has zero
energy, such as at $\vec{Q} = \vec{\tau} \pm \vec{q}_0$. Consequently it was
not possible to determine accurately the energy
widths of the majority of spin waves studied.
Nevertheless it is clear that rather-well-defined
spin waves were observed in spite of the low con-
centration of magnetic ions in the crystals studied.
This is most apparent in the measurements for q's
where only one peak is observed, as shown in Figs.
1(a) and 1(c). The width of each peak in the scat-
tered neutron groups was generally about 0.5 meV
which includes the width of the instrumental resolu-
tion of between 0.3 to 0.4 meV. To determine the
spin-wave energies, groups were fitted by a least-
squares procedure with either Gaussian or Lorentz-
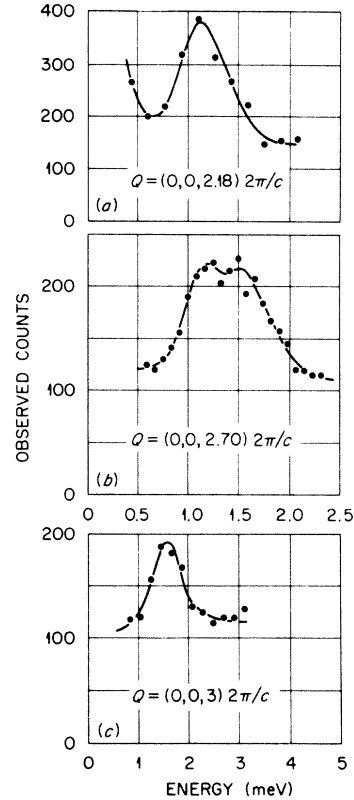ian functions together with a polynomial form of

FIG. 1. Several constant-$\vec{Q}$ scans obtained for
$Y_{0.90}Ho_{0.10}$.

background. The spin-wave dispersion curves ob-
tained from the measurements are shown in Figs.
2 and 3. The experimental uncertainties shown
are largely due to the difficulties associated with
determining the energy of each peak in scans like
Fig. 1(b) where two peaks are present.

III. ANALYSIS AND DISCUSSION

For wave vectors in the c direction, the exchange
function $\mathcal{J}(\vec{q})$ may be written in terms of interplanar
exchange constants[5] $\mathcal{J}_l$ as

$$\mathcal{J}(\vec{q}) = 2(g - 1)^2 \sum_l \mathcal{J}_l \cos(\pi l \, \zeta), \qquad (3)$$

where $|\vec{q}| = \zeta(2\pi/c)$. If one assumes that the inter-
action matrix U_q in Eq. (1) is the same for both
Tb and Ho, the $\mathcal{J}_l$'s may be approximately the
same for the Y(Tb) and the Y(Ho) alloys,
since $\chi(\vec{q})$ for these alloys is expected to be nearly
equal to that of pure yttrium metal. Therefore,
the experimentally observed spin-wave dispersion
curves have been fitted by a least-squares proce-
dure to Eq. (2), with $\mathcal{J}_l$ taken to be common to
both alloys, and the anisotropy constant B as the
fitting parameters. Values of the parameters so
determined are listed in Table I together with their

uncertainties. Seven exchange constants are need-
ed to give a satisfactory fit to the data. A fit with
eight $\mathcal{J}_i$'s did not improve the agreement with the
experimental results. The uncertainty for $\mathcal{J}_2$ is
quite large because in the second bracket on the
right-hand side of Eq. (2) $\mathcal{J}_2$ always occurs in
combination with $\cos(2\pi\zeta_0)$, where $\zeta_0 \cong \vec{q}_0(c/2\pi)$
$= 0.28$. Thus, since $\cos(2\pi\zeta_0)$ is quite small, $2\pi\zeta_0$
being near $\frac{1}{2}\pi$, $\mathcal{J}_2$ contributes very little to the q
dependence of $\hbar\omega(\vec{q})$. A similar problem exists for
$\mathcal{J}_6$. The value obtained for the anisotropy constant
B for Tb is in reasonable agreement with the val-
ues deduced from measurements on pure Tb,[6,7]
whereas that for Ho is larger than expected.[5,7]

The spin-wave dispersion curves calculated with
the parameters given in Table I are shown by the
solid lines in Figs. 2 and 3. The over-all agree-
ment between the experiment and the calculations
is good; however, the experimentally observed
dispersion curve for the Ho alloy seems to have an
initial slope considerably larger than that calculated
from the fit. This discrepancy may be an indica-
tion of the inadequacy of the model used in the
present analysis.

The $\mathcal{J}(\vec{q})$ function calculated from the constants
in Table I is shown in Fig. 4. Since the location
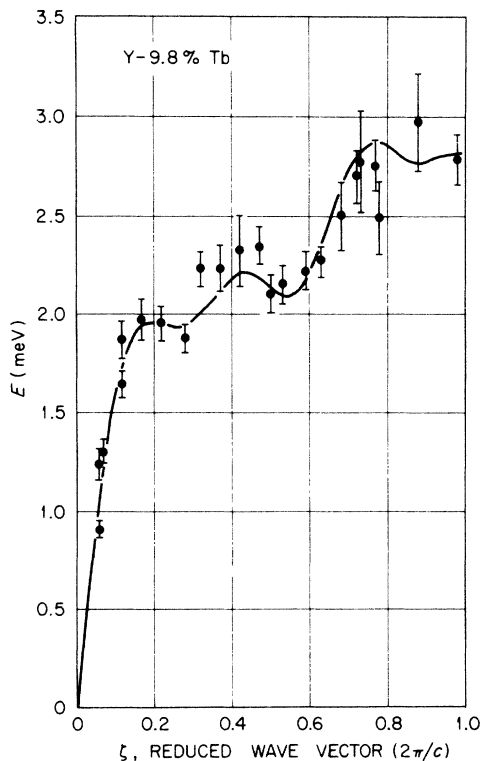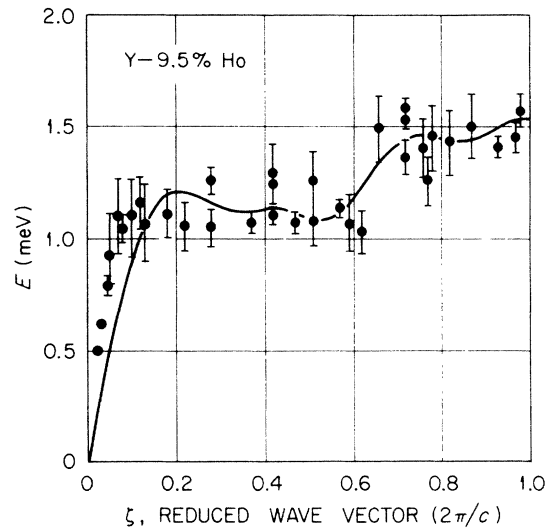of the maximum of $\mathcal{J}(\vec{q})$ determines the periodicity

FIG. 3. Measured spin-wave dispersion relation for
Y—9.5-at.% Ho in the c direction at 4.7 °K. The line
represents a fit to the data as described in the text.

FIG. 2. Measured spin-wave dispersion relation for
Y—9.8-at. % Tb in the c direction at 4.7 °K. The line
represents a fit to the data as described in the text.

of the spiral magnetic structure, one expects $\mathcal{J}(\vec{q})$
to have an absolute maximum at $\vec{q}_0 = 0.28(2\pi/c)$.
Indeed, this condition is nearly fulfilled even
though no restriction was imposed on the fitting
calculation to make $\mathcal{J}(\vec{q}_0)$ the maximum. The mag-
nitude and location of the peak in $\mathcal{J}(\vec{q})$ near 0.28
$\times(2\pi/c)$ is relatively insensitive to the experimental
errors and to the number of exchange constants
$\mathcal{J}_i$ used in fitting the data, whereas the structure
beyond $q \sim 0.5(2\pi/c)$ is extremely sensitive to these
aspects of the data analysis. Thus, although the
existence of the second peak in $\mathcal{J}(\vec{q})$ near $0.6(2\pi/c)$
is quite consistent with the results obtained ex-
perimentally for the rare-earth metals and with
various susceptibility calculations carried out for
these metals,[8] including yttrium, the present re-
sults for $\mathcal{J}(\vec{q})$ at large q are very uncertain.

The $\mathcal{J}(\vec{q})$ obtained for these alloys is quantitatively
quite different from those obtained for the pure
rare-earth metals, as is shown in Fig. 4, where
a comparison with the results for Ho, which has
the largest $\mathcal{J}(\vec{q}_0)$ of the pure metals, is illustrated.
In particular, the peak $\mathcal{J}(\vec{q}_0)$ is much more pro-
nounced in the alloys. This result appears to be
consistent with several other experimental results.
As indicated above, when the pure metals are di-
luted with yttrium the tendency toward helical mag-
netic ordering increases rapidly with increasing
yttrium concentration.[3] For example, the pre-
dominately ferromagnetic behavior of Tb is com-
pletely eliminated at an yttrium concentration of
~ 24 at. %. And the ferromagnetic structure of Dy,
which has a stronger tendency to helical magnetic
order than does Tb, is eliminated at only 5-at.% Y.

TABLE I. Exchange and anisotropy constants for $Y_{0.90}Tb_{0.10}$ and $Y_{0.90}Ho_{0.10}$ alloys. All units are meV.

Exchange constants $\mathscr{J}_i$	
$\mathscr{J}_1$	2.48 ± 0.20
$\mathscr{J}_2$	1.28 ± 1.39
$\mathscr{J}_3$	-0.89 ± 0.20
$\mathscr{J}_4$	-0.93 ± 0.24
$\mathscr{J}_5$	-1.85 ± 0.58
$\mathscr{J}_6$	0.30 ± 0.22
$\mathscr{J}_7$	0.38 ± 0.10

	Anisotropy constants B	
	This work	Results for pure metals
B_{Tb}	0.26 ± 0.04	0.37^a; 0.47^b
B_{Ho}	0.22 ± 0.03	0.02^c; $0.02 - 0.07^d$

[a]Reference 6.

[b]Reference 7.

[c]Reference 5; 50 °K.

[d]Reference 7; large spread exists in results from different laboratories.

In Gd, which is ferromagnetic only, helical ordering occurs in Gd-Y alloys at an Y concentration of ~20 at.%, and ferromagnetism is completely suppressed at ~60-at. % Y. It is tempting to explain such magnetic-structure properties in terms of a concentration-dependent exchange interaction $\mathscr{J}(\vec{q})$ for which $\mathscr{J}(\vec{q}_0) - \mathscr{J}(0)$ and, hence, the stability of the helical structure grows with the yttrium concentration.

Such a model would also explain qualitatively the results of Belov et al.,[2] who find that although the Néel temperatures for Tb_cY_{1-c} alloys decrease as expected with decreasing c, the magnetic-field intensity H_c that is required to transform the helical structure of these alloys to ferromagnetic structures increases strongly with decreasing c for $0.36 \leqslant c \leqslant 1.0$. H_c can be estimated by equating the energy supplied by the field in the ferromagnet, $g\mu_B\sum_i \vec{H} \cdot \vec{J}_i$, to the difference in energy between the spiral and ferromagnetic arrangements, i.e.,

$$g\mu_B \sum_i^{mag} \vec{H} \cdot \vec{J}_i$$

$$\cong \frac{1}{2} \sum_{ij}^{mag} \mathscr{J}_{ij} \exp[i\vec{q}_0 \cdot (\vec{R}_i - \vec{R}_j)] |\vec{J}|^2$$

$$-\frac{1}{2} \sum_{ij}^{mag} \mathscr{J}_{ij} |\vec{J}|^2 ,$$

where μ_B is the Bohr magneton and the summations include only the magnetic ions in the lattice. If the summations over magnetic sites are replaced by summations over all atomic sites by taking into account the probability c that a site is occupied by a magnetic ion, one obtains, with $\vec{H} \| \vec{J}_i$,

$$g\mu_B \sum_i^{mag} \vec{H} \cdot \vec{J}_i \cong g\mu_B H(cJ)$$

and

$$\frac{1}{2} \sum_{ij}^{mag} \mathscr{J}_{ij} \exp[i\vec{q}_0 \cdot (\vec{R}_i - \vec{R}_j)] |\vec{J}|^2 - \frac{1}{2} \sum_{ij}^{mag} \mathscr{J}_{ij} |\vec{J}|^2$$

$$\cong \frac{1}{2} c^2 \mathscr{J}(\vec{q}_0) J(J+1) - \frac{1}{2} c^2 \mathscr{J}(0) J(J+1).$$

Thus

$$g\mu_B H_c J \cong \frac{1}{2} c[\mathscr{J}(\vec{q}_0) - \mathscr{J}(0)] J(J+1).$$

For $c = 0.1$, the H_c as calculated from the results in Fig. 4 is approximately 80 kOe, which may be compared to the ~40–50-kOe field observed by Belov et al. for $c = 0.36$. The agreement is probably reasonable in view of the uncertainties in our $\mathscr{J}(\vec{q}_0) - \mathscr{J}(0)$ results (perhaps ~25%) and the fact that results for rather different concentrations are being compared. Also, we have ignored here the magnetoelastic forces which assist the applied field in stabilizing the ferromagnetic structure, as discussed by Cooper for the cases of Tb and Dy.[9] Thus the field necessary to produce ferromagnetism is expected to be smaller than that predicted by calculations based only on exchange energy considerations.

Recently, theoretical calculations of the conduction-electron susceptibility $\chi(\vec{q})$ have been carried out by Liu et al.[8] for several metals including Y and the heavy rare earths. Their calculations indicate that $\chi(\vec{q}_0) - \chi(0)$ for Y is approximately four times that for Tb. Although the magnitude of the
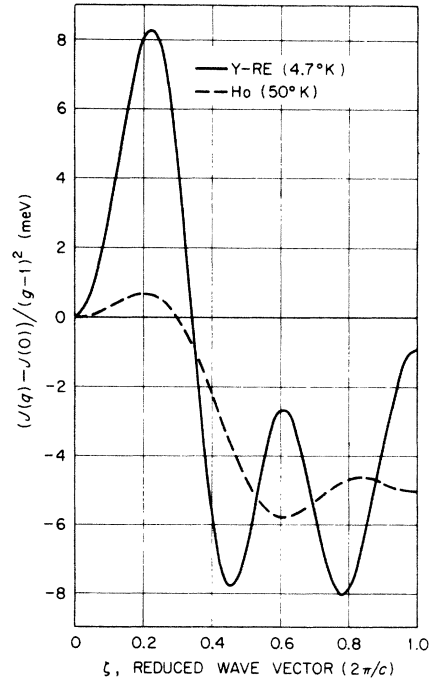

FIG. 4. Exchange interaction $[\mathscr{J}(\vec{q}) - \mathscr{J}(0)]/(g-1)^2$ deduced from the measured spin-wave dispersion relations for the $Y_{0.90}Tb_{0.10}$ and $Y_{0.90}Ho_{0.10}$ alloys and for Ho.

exchange interaction U_q in Eq. (1) is not known quantitatively, this theoretical result for $\chi(\vec{q})$ is at least qualitatively consistent with our $\mathcal{J}(\vec{q})$ results.

One consequence of the mean-lattice assumption employed here is that single-ion interactions, such as the crystal field, are independent of c, whereas two-ion interactions, such as the exchange, are proportional to c. In the limit of low concentrations the crystal-field contribution to the magnetic Hamiltonian can become comparable to that of the exchange interaction. Therefore the magnetic ground state of these alloys may not correspond to the pure (only exchange) eigenstate $|J_z=J\rangle$ as was implicitly assumed in the conventional spin-wave theory we used. In the absence of exchange, the ground state for both Tb ($J=6$) and Ho ($J=8$) has $J_z=0$. To investigate this problem the single-ion levels appropriate to Eq. (A1) in a mean-field approximation have been calculated, and the collective excitations based on these levels have been investigated as a possible alternative description for the present experimental data. The details of these calculations will be described in a future publication.

The dielectric function which describes the screening of the ionic motion by the electrons and governs the electronic contributions to the dynamical matrix also depends on $\chi(q)$. Thus Kohn-type anomalies or kinks can occur in the phonon dispersion curves owing to peaks in $\chi(\vec{q})$. A thorough search for such anomalies has been carried out for both alloys. Although structure has been found in the shapes of several phonon branches, the observed structure appears to be related to resonant phonon modes associated with the heavy Tb or Ho "mass impurities" in the Y structure and not to the peaks in $\chi(\vec{q})$. A detailed account of this phonon investigation will be given in a separate publication.

APPENDIX

The Hamiltonian for the magnetic ions is assumed to consist of the following terms:

$$\mathcal{H}= B \sum_i^{mag} S_{i\zeta}^2 -\frac{1}{2} \sum_{ij}^{mag} J_{ij} \vec{S}_i \cdot \vec{S}_j, \qquad (A1)$$
$$i \neq j$$

where B is a crystal-field anisotropy parameter, J_{ij} is the exchange interaction between rare-earth ions having total angular momenta $\vec{S}_i$ and $\vec{S}_j$, and the ζ direction is in the c direction. As indicated, the summations include only the magnetic ions in the lattice. The usual transformation to a rotating coordinate system, such that the equilibrium direction of the moment at each site determines the z direction at that site, is made,[6] so that

$$\mathcal{H}=\frac{1}{4} B \sum_i^{mag} (S_i^+ S_i^+ + S_i^- S_i^- + 2 S_i^- S_i^+)$$

$$-\frac{1}{2} \sum_{i \neq j}^{mag} \sum^{mag} J_{ij} [\frac{1}{4}(1-\cos\phi_{ij})(S_i^+ S_j^+ + S_i^- S_j^-)$$

$$+\frac{1}{4}(1+\cos\phi_{ij})(S_i^+ S_j^- + S_i^- S_j^+)$$

$$-\frac{1}{2}\cos\phi_{ij}(S_i^+ S_i^- + S_j^+ S_j^-)], \qquad (A2)$$

where the standard replacement of S_x and S_y by the appropriate combinations of S^+ and S^- has been used; and where $\phi_{ij}=\phi_i-\phi_j$ with $\phi_i=\vec{Q}\cdot\vec{R}_i$, where $\vec{Q}$ is the wave vector of the static spiral structure. For operators that vary in time as $e^{i\omega t}$, one has for the equations of motion

$$i\hbar \dot{S}_i^+ = -\hbar\omega S_i^+ = [S_i^+, \mathcal{H}], \qquad (A3)$$

$$i\hbar \dot{S}_i^- = -\hbar\omega S_i^- = [S_i^-, \mathcal{H}]. \qquad (A4)$$

For example, from Eqs. (A2) and (A3) we obtain

$$-\hbar\omega S_i^+ = BS(S_i^+ + S_i^-) - \frac{1}{2} S \sum_{i, l \neq i}^{mag} J_{il}[(1-\cos\phi_{il})S_i^-$$

$$+(1+\cos\phi_{il})S_i^+ - 2\cos\phi_{il} S_i^+]. \qquad (A5)$$

We then multiply both sides of Eq. (A5) by $\exp \times(i\vec{q}\cdot\vec{R}_i)$; sum over l, the sites of the magnetic ions; and make the following assumptions concerning the average $\langle\cdots\rangle$ of the resulting equation over all configurations of the random lattice:

$$\left\langle \sum_i^{mag} S_i^\pm \exp(\pm i\vec{q}\cdot\vec{R}_i) \right\rangle \simeq c \sum_l^{all} S_l^\pm \exp(\pm i\vec{q}\cdot\vec{R}_i) = c S_q^\pm$$

and

$$\left\langle \sum_{i, l\, i\neq l}^{mag} J_{il}(1-\cos\phi_{il})S_i^- \exp(i\vec{q}\cdot\vec{R}_i) \right\rangle$$

$$=\left\langle \sum_{i, l\, i\neq l}^{mag} J_{il}(1-\cos\phi_{il}) \right.$$

$$\left. \exp(-i\vec{q}\cdot\vec{R}_{il})S_i^-\exp(i\vec{q}\cdot\vec{R}_i) \right\rangle$$

$$\simeq c^2 [J(\vec{q}) - \frac{1}{2}J(\vec{Q}+\vec{q}) - \frac{1}{2}J(\vec{Q}-\vec{q})] S_{-q}^-,$$

where $\vec{R}_{il}=\vec{R}_i-\vec{R}_l$ and c is the concentration of magnetic ions in the lattice. This procedure leads to

$$-\hbar\omega S_q^+ = BS(S_q^+ + S_{-q}^-) - \frac{1}{2}cS[J(\vec{q}) - \frac{1}{2}J(\vec{Q}+\vec{q})$$

$$-\frac{1}{2}J(\vec{Q}-\vec{q})]S_{-q}^- - \frac{1}{2}cS[J(\vec{q}) + \frac{1}{2}J(\vec{Q}+\vec{q}) + \frac{1}{2}J(\vec{Q}-\vec{q})]$$

$$\times S_q^+ + cS J(\vec{Q}) S_q^+. \qquad (A6)$$

From (A4) a similar equation for $-\hbar\omega S_{-q}^-$ can be obtained to give the usual set of simultaneous equations and eigenvalue calculation, viz.,

$$(A_q + \hbar\omega) S_q^+ + B_q S_{-q}^- = 0, \qquad (A7)$$

$$-B_q S_q^+ + (-A_q + \hbar\omega) S_{-q}^- = 0,$$

with the solution

$$\hbar\omega = [(A_q - B_q)(A_q + B_q)]^{1/2}$$

or

$$\hbar\omega = S \left(c[\, J(\vec{Q}) - \tfrac{1}{2}J(\vec{Q}+\vec{q}) - \tfrac{1}{2}J(\vec{Q}-\vec{q})]\right)$$

$$\times \{c[\, J(\vec{Q}) - J(\vec{q})] + 2\,B\}\}^{1/2} .$$

This equation is the same as Eq. (2) with the obvious change in notation $\vec{Q} \to \vec{q}_0$ and $S \to J$.

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