

Diluted quasi-one-dimensional classical antiferromagnets

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We consider the effect of the exchange disordering, mainly the nonmagnetic dilution, on the low-temperature properties of the quasi-one-dimensional classical antiferromagnets. Dynamic susceptibility expansion in defect concentration shows that even a low level of doping substantially changes the spin-wave stiffness and the transverse susceptibility. We investigate the nonperturbative concentration dependencies of the spin-wave velocities and susceptibility arising when the impurity concentration exceeds the small characteristic value $x_0 = \sqrt{J_{\perp}/J_{\parallel}}$. A general result, pertinent to the true three-dimensional antiferromagnets as well, concerns the excitation spectrum of the disordered nonfrustrated antiferromagnets with nonfluctuating anisotropy. In this case the hydrodynamical description of long-wave excitations breaks down, and the spin waves with energies near the anisotropy gap are overdamped.

I. INTRODUCTION

The properties of the one-dimensional (1D) and quasi-one-dimensional antiferromagnets have been intensively investigated for many years. It is well known that there is no long-range magnetic order in the ground state of the true one-dimensional antiferromagnet. The (small) interchain interaction J_{\perp} , which is always present in any quasi-one-dimensional substance, suppresses the strong quantum spin fluctuations in the chains and leads to a three-dimensional magnetic order. This effect of J_{\perp} differs for different spin systems. For integer spin- S systems, the J_{\perp} coupling should exceed a threshold value to be able to produce an ordered state,¹ while for half-integer spin- S systems antiferromagnetism is established whatever the small J_{\perp} is. Clearly, the properties of the quasi-one-dimensional antiferromagnets characterized by strong zero-point spin fluctuations can be quite unusual.

The spin-wave expansion of staggered magnetization demonstrates that these fluctuations are suppressed only if the parameter $\gamma = (2\pi S)^{-1} \ln(J_{\parallel}/J_{\perp})$ is small (J_{\parallel} is the in-chain coupling). The corresponding system may be treated classically. Let us stress that the parameter γ depends on J_{\parallel}/J_{\perp} only logarithmically, while the prelogarithmic factor is small for $S > \frac{1}{2}$. This explains why the Néel ground state is a good approximation for many strongly anisotropic antiferromagnetic systems.^{2,3}

In this paper we consider the effects of disorder in the classical quasi-1D antiferromagnets. The accent is put on the disorder resulting from nonmagnetic dilution.

It was recognized quite long ago that nonmagnetic impurities cut the chains into segments, connected only by a weak interchain coupling and, thus, strongly affect their magnetic properties. There was a series of very illuminative experimental and theoretical papers (Refs. 4–7 and

references therein) in the 1970s and 1980s that focused on the study of the Néel temperature dependence of the dopant atom concentration. It was shown, e.g., that 0.5% of Cd atoms depress the Néel temperature of $(\text{CH}_3)_4\text{NMnCl}_3$ (TMMC) by a factor of 2. The theory derived in Ref. 6, although not rigorous, gives a fairly satisfactory interpretation of these measurements.

The available experimental and theoretical results are concerned mainly with the situation above the Néel temperature. As for the low-temperature static and dynamical properties, the information is poor and fragmentary. Neither systematic measurements of the transversal spin susceptibility and spin-wave spectrum has been performed nor a theory for the concentration dependence of these quantities been worked out. We are aware only of the very interesting neutron studies of static and dynamic spin correlations inside one-dimensional segments.⁷ However, no data have been obtained concerning the spectrum of the spin waves delocalized due to segment exchange coupling via a J_{\perp} interaction.

We would like to point out that it is practically impossible to get such data on materials such as TMMC with low Néel temperatures $T_N \approx 1$ K. As we will see below, the upper limit of the delocalized spin-wave excitation spectrum $\omega_{\max} < T_N$ ($x=0$) is too small to be resolved in a neutron-inelastic-scattering experiment (x is the nonmagnetic impurity concentration). From this point of view, magnets such as $\text{K}_2\text{Fe}_{1-x}\text{Ga}_x\text{F}_5$, where $T(x=0) = 6.95$ K,⁸ look more promising.

Here we present a theory of transverse magnetic susceptibility and the spin-wave spectrum of disordered classical quasi-1D antiferromagnets. The concentration expansion of the dynamical transverse susceptibility demonstrated to us that in this case one should expand in $x(J_{\parallel}/J_{\perp})^{1/2}$. It follows that only at $x \ll x_0 = (J_{\perp}/J_{\parallel})^{1/2}$ are the concentration corrections to all the magnetic

characteristics small. If $x_0 \ll x \ll 1$, the susceptibility χ strongly depends on x , and this dependence cannot be calculated perturbatively. This is in contrast to the case of the isotropic system with $J_{\perp} = J_{\parallel}$, where the natural expansion parameter is x .

We will see that in the concentration interval $x_0 \ll x \ll 1$ the excitations inside the segment can be neglected. Thus we can consider segments as rigid spin molecules connected by the J_{\perp} coupling. This gives for the static transversal susceptibility $\chi \propto x^2 \ln x^{-1}$. The transversal and longitudinal spin-wave velocities are proportional correspondingly to $x^{-2} (\ln x^{-1})^{-1/2}$ and $x^{-1} (\ln x^{-1})^{-1/2}$. The system becomes more isotropic as x increases.

As a by-product of our calculations, we received a general result concerning the spin-wave spectrum of any antiferromagnetic system with exchange disorder. It is known that in a Heisenberg system low-frequency spin waves are hydrodynamical however strong the disorder is, provided there is no frustration. In this paper we show that a combination of small nonfluctuating easy-axis magnetic anisotropy and nonfrustrated exchange disorder leads to nonhydrodynamical spin-wave behavior: The spin waves are overdamped in the $q \rightarrow 0$ limit.

Clearly, nothing like this can occur in ferromagnets. Indeed, while no solution with uniform rotations of sublattices is possible for disordered antiferromagnets, for ferromagnets a uniform spin rotation *does* give the $q \rightarrow 0$ solution. Our result may explain the experimental observation by Uemura and Birgeneau.⁹

The paper is organized as follows. In Secs. II and III we present the results of a concentration expansion for bond and site defect models. In Sec. II one can find also a discussion of nonhydrodynamic spin-wave behavior in easy-axis antiferromagnets with exchange disorder. The nonperturbative case $x > x_0$ is discussed in Sec. IV.

A short version of this paper has been published in Ref. 10.

II. VERY DILUTED SYSTEMS: BOND PROBLEM

To begin with, let us consider the simplest defect: a removed bond between two nearest neighbors in a chain. In the experiments mentioned above, the site-diluted antiferromagnets have been investigated, but the simplest model we are going to study in this section allows us to understand the most important features of the problem in the low-concentration limit.

The Hamiltonian of a two-sublattice quasi-1D antiferromagnet with a removed bond between the nearest-neighbor spins \mathbf{S}_m and \mathbf{S}_n in the chain may be written as

$$H = \sum_{\langle ij \rangle} J_{ij} (\mathbf{S}_{i1} \cdot \mathbf{S}_{j2}) - \Delta \sum_{p=1,2;i} (S_{ip}^z)^2 - J_{\parallel} (\mathbf{S}_{m1} \cdot \mathbf{S}_{n2}). \quad (1)$$

Here the sum is over all nearest-neighbor bonds, $\Delta > 0$ is the magnetic anisotropy constant, and the intrachain J_{\parallel} and interchain J_{\perp} exchange interaction energies are positive.

We shall consider low temperatures $T \ll T_N \sim (J_{\parallel} J_{\perp})^{1/2}$, when the thermal fluctuations can be neglected, and suppose that the parameter $\gamma \ll 1$. Thus, as has been explained in the Introduction, the quantum fluctuations can be neglected too. Let us introduce the retarded boson Green's function

$$G_{ij}^{pp'}(t-t') = -i\Theta(t-t') \langle [c_{ip}(t), c_{jp'}^{\dagger}(t')] \rangle_T, \quad (2)$$

where $\Theta(t)$ is the step function, $\langle \dots \rangle_T$ denotes a Boltzmann average, and c_{ip}, c_{ip}^{\dagger} are the Holstein-Primakoff operators for the spins in the sublattice p .

The transformation from \mathbf{S} to c, c^{\dagger} is

$$\begin{aligned} S_{i1}^+ &= (2S)^{1/2} c_{i1}, & S_{i1}^- &= (2S)^{1/2} c_{i1}^{\dagger}, & S_{i1}^z &= S - c_{i1}^{\dagger} c_{i1}, \\ S_{i2}^+ &= (2S)^{1/2} c_{i2}^{\dagger}, & S_{i2}^- &= (2S)^{1/2} c_{i2}, & S_{i2}^z &= -S + c_{i2}^{\dagger} c_{i2}. \end{aligned} \quad (3)$$

In an ordered crystal the Green's-function matrix is given by

$$\hat{G}_0(\omega, \mathbf{q}) = g(\omega, \mathbf{q}) \begin{pmatrix} \omega + J(0)S + 2S\Delta & -SJ(\mathbf{q}) \\ -SJ(\mathbf{q}) & -\omega + J(0)S + 2S\Delta \end{pmatrix}, \quad (4)$$

where

$$g(\omega, \mathbf{q}) = (\omega^2 - \omega_{\mathbf{q}}^2)^{-1}. \quad (5)$$

The spin-wave spectrum

$$\omega_{\mathbf{q}}^2 = S^2 \{ [J(0) + 2\Delta]^2 - J^2(\mathbf{q}) \}. \quad (6)$$

In the quasi-1D antiferromagnets, the spin-wave dispersion law can be written in the form

$$\begin{aligned} \omega^2(\mathbf{q}) &= \omega_{\parallel}^2(q_z) + \omega_{\perp}^2(\mathbf{q}_{\perp}) + 4J_{\parallel}(0)S^2\Delta, \\ \omega_{\parallel}^2(q_z) &= S^2 [J_{\parallel}^2(0) - J_{\parallel}^2(q_z)], \\ \omega_{\perp}^2(\mathbf{q}_{\perp}) &= 2S^2 J_{\parallel}(0) [J_{\perp}(0) - J_{\perp}(\mathbf{q}_{\perp})]. \end{aligned} \quad (7)$$

The axis z is directed along the chains and $\mathbf{q}_{\perp} = (0, q_x, q_y)$. At small q and $\Delta = 0$, the spectrum is linear with velocities

$$c_{0\parallel} \sim J_{\parallel}(0)S, \quad c_{0\perp} \sim [J_{\parallel}(0)J_{\perp}(0)]^{1/2}S.$$

The Green's function in the doped crystal can be writ-

ten in the form

$$\hat{G}(\mathbf{q}, \mathbf{q}_1) = \hat{G}_0(\mathbf{q})\delta_{\mathbf{q}, \mathbf{q}_1} + \hat{G}_0(\mathbf{q})\hat{T}(\mathbf{q}, \mathbf{q}_1)\hat{G}_0(\mathbf{q}_1), \quad (8)$$

where the single-defect T matrix is governed by the equation

$$\hat{V}(\mathbf{q}, \mathbf{q}_1) = -2J_{\parallel}S \begin{bmatrix} \exp[i(\mathbf{q}-\mathbf{q}_1)\cdot\mathbf{R}_m] & \exp(i\mathbf{q}\cdot\mathbf{R}_m - i\mathbf{q}_1\cdot\mathbf{R}_n) \\ \exp(i\mathbf{q}\cdot\mathbf{R}_n - i\mathbf{q}_1\cdot\mathbf{R}_m) & \exp[i(\mathbf{q}-\mathbf{q}_1)\cdot\mathbf{R}_n] \end{bmatrix}. \quad (10)$$

The integral Eq. (8) can be solved easily, since all matrix elements of the perturbation energy $V(\mathbf{q}, \mathbf{q}_1)$ are separable functions. The solution can be written as

$$\hat{T}(\mathbf{q}, \mathbf{q}_1) = \Lambda^{-1}(\omega)\hat{V}(\mathbf{q}, \mathbf{q}_1), \quad (11)$$

where

$$\Lambda(\omega) = 1 + \frac{2J_{\parallel}S^2}{N} \sum_{\mathbf{q}} [J(0) - J(\mathbf{q}) \cos q_z d_{\parallel} + 2\Delta] g(\omega, \mathbf{q}); \quad (12)$$

d_{\parallel} is the lattice constant in the z direction.

One can easily check (and we will show it a little bit later) that for quasi-1D systems $\Lambda(0) \sim (J_{\perp}/J_{\parallel})^{1/2} \ll 1$. Thus, at low frequencies, the scattering by the defects is strongly enhanced.

Averaging Eq. (7) over the random distribution of the removed bonds, we obtain at first order in the defect concentration x the following expression for the averaged function $\hat{G}(\omega, \mathbf{q})$:

$$\hat{G}(\omega, \mathbf{q}) = \hat{G}_0(\omega, \mathbf{q}) + \hat{G}_0(\omega, \mathbf{q})x\hat{T}(\mathbf{q}, \mathbf{q})\hat{G}_0(\omega, \mathbf{q}). \quad (13)$$

The dynamical susceptibility

$$\chi(\omega, \mathbf{q}) = -S \sum_{pp'} G^{pp'}(\omega, \mathbf{q}) \quad (14)$$

can be now calculated immediately. It follows from Eqs. (9)–(13) that

$$\chi(\omega, \mathbf{q}) = \frac{2S^2[J(0) + 2\Delta - J(\mathbf{q}) - xJ_{\parallel}(1 - \cos q_z d_{\parallel})\Lambda^{-1}(\omega)]}{\omega^2 - \omega_q^2 - 2x\omega_q \Sigma(\omega, \mathbf{q})}. \quad (15)$$

Here the self-energy $\Sigma(\omega, \mathbf{q})$ equals

$$\Sigma(\omega, \mathbf{q}) = -\Lambda^{-1}(\omega)\omega_q^{-1}J_{\parallel}S^2[J(0) + 2\Delta - J(\mathbf{q}) \cos q_z d_{\parallel}]. \quad (16)$$

For the Heisenberg antiferromagnets ($\Delta=0$), the spin-wave dispersion is given by the equation

$$\omega^2 = \omega^2(\mathbf{q}) \{ 1 - x\Lambda^{-1}(\omega)\omega^{-2}(\mathbf{q})[\omega_{\parallel}^2(\mathbf{q}) + \frac{1}{2}\omega_{\perp}^2(\mathbf{q})] \}. \quad (17)$$

In the hydrodynamic limit $\omega \ll S[J_{\perp}(0)J_{\parallel}(0)]^{1/2}$, the renormalization of the spin-wave velocity is proportional

$$\hat{T}(\mathbf{q}, \mathbf{q}_1) = \hat{V}(\mathbf{q}, \mathbf{q}_1) + \frac{1}{N} \sum_{\mathbf{q}_2} \hat{V}(\mathbf{q}, \mathbf{q}_2)\hat{G}_0(\mathbf{q}_2)\hat{T}(\mathbf{q}_2, \mathbf{q}_1). \quad (9)$$

Here $\hat{V}(\mathbf{q}, \mathbf{q}_1)$ is the perturbation energy and N is the number of sites in a sublattice. For the model described by the Hamiltonian (1), the matrix $\hat{V}(\mathbf{q}, \mathbf{q}_1)$ has the form

to $\Lambda^{-1}(0)$. For quasi-1D systems, $\Lambda(0)$ can be written as

$$\Lambda(0) = \frac{\Omega}{(2\pi)^3} \int d^3q \frac{J_{\perp}(0)J_{\parallel}(0) - J_{\perp}(\mathbf{q})J_{\parallel}(\mathbf{q})}{J^2(0) - J^2(\mathbf{q})}, \quad (18)$$

where Ω is the unit-cell volume.

The main contribution to the integral (18) comes from the small values of the longitudinal component of the momentum $q_z \sim (J_{\perp}/J_{\parallel})^{1/2}$. Thus the denominator in Eq. (18) can be expanded in q_z^2 . Integrating over q_z , we find

$$\Lambda(0) = \alpha [J_{\perp}(0)/8J_{\parallel}(0)]^{1/2}, \quad (19)$$

$$\alpha = \frac{\Omega}{(2\pi)^2} \int d^2q_{\perp} \left[1 - \frac{J_{\perp}(\mathbf{q}_{\perp})}{J_{\perp}(0)} \right]^{1/2}.$$

Equations (15)–(19) yield the renormalization of the spin-wave velocity,

$$c(\theta) - c_0(\theta) = -x\alpha^{-1}(8J_{\parallel}/J_{\perp})^{1/2}(c_{0\parallel} \cos^2\theta + \frac{1}{2}c_{0\perp} \sin^2\theta), \quad (20)$$

where $c(\theta)$ [$c_0(\theta)$] is the spin-wave velocity in the doped [undoped] crystal and θ denotes the angle between \mathbf{q} and the z direction.

One can see from Eq. (20) that the concentration expansion parameter is not x , but rather $x(J_{\parallel}/J_{\perp})^{1/2}$; i.e., the modification of the spectrum by impurities is large even at small x if $x > x_0 = (J_{\perp}/J_{\parallel})^{1/2}$. At such concentrations the expansion in a power series in x is not applicable, and new concentration dependences of the spin-wave velocity and susceptibility should be expected.

To clarify the origin of the parameter $x(J_{\parallel}/J_{\perp})^{1/2}$, let us consider the motion of the spin excitation near the defect. We denote by n the number of transversal bonds, which the spin excitation uses to go around the defect. Then the time τ that the spin excitation needs to go around the defect is a sum of $1/J_{\perp}n$ and n/J_{\parallel} :

$$\tau \simeq \frac{n}{J_{\parallel}} + \frac{1}{J_{\perp}n}. \quad (21)$$

Indeed, the term $1/J_{\perp}n$ is a characteristic time of jumping to an adjacent chain, while the term n/J_{\parallel} describes the time necessary to go through the chain section containing n transversal bonds.

It is not accidental that Eq. (21) looks like the formula

for the connection of conductivities J_{\parallel} and J_{\perp} in parallel and in series. The deep analogy between the propagation of spin excitations in an antiferromagnet with exchange couplings J_{ij} and electrical current in the network with conductances $\sigma_{ij} \propto J_{ij}$ between the sites i and j has been established by Harris and Kirkpatrick.¹¹ The time τ has a minimum (the probability to leave the chain has a maximum) for large $n = (J_{\parallel}/J_{\perp})^{1/2}$. Perturbation theory breaks down when this quantity is of the order of the mean segment length x^{-1} .

The spin-wave damping is given by

$$\begin{aligned} \Gamma(\omega, \mathbf{q}) &= \frac{x}{2\omega_{\mathbf{q}}} \text{Im}\Sigma(\omega, \mathbf{q}) \\ &= \frac{x\Omega J_{\parallel}(0)\omega^3(c_{0\parallel}^2 q_z^2 + 2^{-1}c_{0\perp}^2 q_{\perp}^2)}{3\pi J_{\perp}(0)c_{0\parallel}c_{0\perp}^2 \alpha^2 \omega(\mathbf{q})}. \end{aligned} \quad (22)$$

The relative damping $\Gamma(\omega, \mathbf{q})/\omega_{\mathbf{q}} \ll 1$ if $\omega \ll S(J_{\perp}J_{\parallel})^{1/2}$ and $x \ll x_0$.

When the frequency exceeds the maximum transversal spin-wave frequency $\omega \gg S(J_{\parallel}J_{\perp})^{1/2}$, the transversal interaction can be omitted in Eq. (16). Thus

$$\Lambda(\omega) = i \frac{\omega}{4J_{\parallel}(0)} + O(\omega^2/J_{\parallel}^2)$$

and

$$\omega^2 = \omega_{\parallel}^2(q_z) + i4x\omega_{\parallel}(q_z)J_{\parallel}(0). \quad (23)$$

If the spin-wave length $\lambda = q_z^{-1}$ is smaller than the mean segment length x^{-1} , the second term on the right-hand side of the Eq. (23) is small; i.e., well-defined spin excitations within a segment exist.

The uniform static susceptibility obtained from Eqs. (15), (16), and (19) is given by

$$\chi = J_{\parallel}^{-1}(0) \{ 1 + x\alpha [2J_{\parallel}(0)/J_{\perp}(0)]^{1/2} \}. \quad (24)$$

The susceptibility increases rapidly with the increase of the doping. At small defect concentration $x \approx x_0$, the susceptibility is approximately 2 times larger than in the undoped crystal.

Let us consider the effect of a small magnetocrystalline anisotropy $\Delta \ll J_{\perp}(0)$. It follows from Eq. (16) that at $q \rightarrow 0$ the self-energy is given by

$$\begin{aligned} \Sigma(\omega - \omega_0, \mathbf{q}) &= \omega_{\mathbf{q}}^{-1} J_{\parallel}(0) S^2 \Delta \Lambda^{-1}(\omega_0) \\ &= \frac{1}{2} \omega_0 \left[\alpha \left[\frac{2J_{\parallel}(0)}{J_{\perp}(0)} \right]^{1/2} \right. \\ &\quad \left. + i2\Omega S^2 \alpha^2 \pi^{-2} \frac{[2\omega_0(\omega - \omega_0)]^{1/2}}{J_{\perp}(0)c_{0\perp}^2 c_{0\parallel}} \right], \end{aligned} \quad (25)$$

where $\omega_0 = 2S[J_{\parallel}(0)\Delta]^{1/2}$.

Thus the renormalization of the spin-wave gap is

$$\omega(q=0, x=0) - \omega(q=0, x) = x\alpha\omega_0 [J_{\parallel}(0)/2J_{\perp}(0)]^{1/2}. \quad (26)$$

It is proportional to the same parameter $(J_{\parallel}/J_{\perp})^{1/2}$ as the correction to the spin-wave velocity. The relative damping

$$\frac{\text{Im}\omega}{\omega - \omega_0} \sim x \left[\frac{\omega_0}{\omega - \omega_0} \right]^{1/2} \frac{\Delta^{3/2}}{J_{\perp} J_{\parallel}^{1/2}} \quad (27)$$

becomes large at small $q \propto (\omega - \omega_0)^{1/2}$. This means that the hydrodynamic description of spin-wave excitations breaks down, and the spin-wave scattering resembles the scattering of electrons on a short-range random potential. This result does not depend on the relation between J_{\parallel} and J_{\perp} and is valid for any disordered antiferromagnet. Indeed, the equations of motion for S_{i1}^+ and S_{i2}^+ in a disordered antiferromagnet can be written as

$$\begin{aligned} \omega S_{i1}^+ &= S \sum_k J_{ik} S_{i1}^+ + S \sum_k J_{ik} S_{k2}^+ + 2\Delta S S_{i1}^+, \\ \omega S_{i2}^+ &= -S \sum_k J_{ik} S_{i2}^+ - S \sum_k J_{ik} S_{i1}^+ - 2\Delta S S_{i2}^+. \end{aligned} \quad (28)$$

It is easy to verify that the uniform rotation of the sublattices $S_{i1}^+ = C_1$, $S_{i2}^+ = C_2$ does not satisfy these equations.

Let us recall that in disordered ferromagnets the situation is quite different. The equations of motion for S_i^+ have a uniform solution with a shifted frequency $\omega - 2\Delta S$; i.e., the anisotropy does not affect the spin-wave damping at all.

It should be noted that an anomalously strong increase of the spin-wave damping has been observed in a diluted antiferromagnet $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$ when x was near the percolation threshold, i.e., when the exchange disorder was sufficiently high.⁹

III. SITE DEFECTS

The problem of spin-wave renormalization in a cubic crystal with site defects was studied many years ago. A comprehensive review can be found in Refs. 12 and 13. Here we shall consider how these results are modified by strong exchange anisotropy in a tetragonal antiferromagnet.

Suppose that the host spin in a site \mathbf{R} is substituted by an impurity spin S' , interacting with nearest neighbors by the exchange energy $J'(\mathbf{r})$. The Hamiltonian can be written as the sum of the perfect crystal Hamiltonian and the interaction Hamiltonian H_{int} , which equals

$$H_{\text{int}} = \sum_{\delta} [J'(\delta) \mathbf{S}'_{\mathbf{R}} \cdot \mathbf{S}_{\mathbf{R}+\delta} - J(\delta) \mathbf{S}_{\mathbf{R}} \cdot \mathbf{S}_{\mathbf{R}+\delta}]. \quad (29)$$

The vector δ points to the nearest neighbors of the impurity spin S' .

According to Wan, Harris, and Kumar,¹² it is convenient to define the operators c_{ip}, c_{ip}^{\dagger} on the defect site via a non-Hermitian transformation

$$\begin{aligned} S_{i1}^+ &= (2S)^{1/2} c_{i1}, \\ S_{i1}^- &= (2S)^{1/2} c_{i1}^{\dagger} [1 + (\beta - 1)\phi_i], \\ S_{i2}^+ &= (2S)^{1/2} c_{i2}^{\dagger}, \\ S_{i2}^- &= (2S)^{1/2} c_{i2} [1 + (\beta - 1)\phi_i], \end{aligned} \quad (30)$$

where $\beta = S'/S$; $\phi_i = 1$, if site i is occupied by the impurity and $\phi_i = 0$ otherwise.

The Hamiltonian (29) can be rewritten with the aid of (30) in terms of the operators c_{ip} and c_{ip}^\dagger

$$H_{\text{int}} = S \sum_{\delta} (c_{R1}^\dagger + c_{R+\delta 2}) \{ c_{R1} [J'(\delta) - J(\delta)] + c_{R+\delta 2} [J'(\delta)\beta - J(\delta)] \}. \quad (31)$$

It is seen from this expression that the interaction matrix for the simple tetragonal lattice is a 7×7 matrix with elements depending on four exchange parameters

$$\hat{U} = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 1/\sqrt{2} & 0 & 0 & 1/2 \\ 0 & 1/2 & 0 & -1/\sqrt{2} & 0 & 0 & 1/2 \\ 0 & 1/2 & 0 & 0 & 1/\sqrt{2} & 0 & -1/2 \\ 0 & 1/2 & 0 & 0 & -1/\sqrt{2} & 0 & -1/2 \\ 0 & 0 & 1/\sqrt{2} & 0 & 0 & 1/\sqrt{2} & 0 \\ 0 & 0 & 1/\sqrt{2} & 0 & 0 & -1/\sqrt{2} & 0 \end{pmatrix}. \quad (33)$$

After the matrix $T^{(\mu)}$ is found (μ labels the representations), the Green's function, averaged over the impurity distribution, can be obtained at first order in the defect concentration from Eq. (13) with

$$\hat{T}(\mathbf{q}, \mathbf{q}_1) = \sum_{\mu} \hat{T}^{\mu}(\mathbf{q}, \mathbf{q}_1). \quad (34)$$

\hat{T} is a 2×2 matrix in the sublattice space.

The self-energy $\Sigma(\omega, \mathbf{q})$, defined by

$$\omega^2 = \omega_q^2 + 2x\omega_q \Sigma(\omega, \mathbf{q}), \quad (35)$$

can be written in terms of the T -matrix elements

$$\Sigma(\omega, \mathbf{q}) = (2\omega_q)^{-1} [J(0)S(T_{11} + T_{22}) - J(\mathbf{q})S(T_{12} + T_{21}) + \omega(T_{11} - T_{22})]. \quad (36)$$

For a quasi-1D crystal, only the representations A_{1g} [the first three columns in the matrix (33)] and A_{2U} [the fifth column in (33)] are relevant, since only these representations describe the spin-wave scattering by the perturbations of the in-chain interaction proportional to ϵ_{\parallel} and ρ_{\parallel} .

Let us begin with A_{2U} . It follows from Eqs. (32) and (34) that the corresponding perturbation energy is (for definiteness we put the impurity at a site of sublattice 1)

$$V_1^{(2)} = 2\rho_{\parallel} S \frac{1}{N^2} \sum_{\mathbf{q}, \mathbf{q}'} \sin(q_z d_{\parallel}) \sin(q'_z d_{\parallel}) c_{\mathbf{q}2} c_{\mathbf{q}'2}^\dagger. \quad (37)$$

Equations (9) and (37) yield

$$T_1^{(2)}(\mathbf{q}, \mathbf{q}') = \frac{2\rho_{\parallel} S}{\Lambda^{(2)}(\omega)} \sin(q_z d_{\parallel}) \sin(q'_z d_{\parallel}), \quad (38)$$

where

$$\begin{aligned} \epsilon_{\parallel} &= J'_{\parallel} - J_{\parallel}, & \epsilon_{\perp} &= J'_{\perp} - J_{\perp}, \\ \rho_{\parallel} &= J'_{\parallel} \beta - J_{\parallel}, & \rho_{\perp} &= J'_{\perp} \beta - J_{\perp}. \end{aligned} \quad (32)$$

Here J'_{\parallel} and J'_{\perp} are the nearest-neighbor interaction energies along and perpendicular to the tetragonal axis. The numbers of the nearest neighbors are $z_{\parallel} = 2$, $z_{\perp} = 4$.

To solve the set of Eqs. (8) and (9), we should use the unitary transformation, decoupling the set of equations into subsets in accordance with the irreducible representations of the crystal point group. For the simple tetragonal crystal, the matrix of the transformation is¹³

$$\Lambda^{(2)}(\omega) = 1 - 2\rho_{\parallel} S \frac{1}{N} \sum_{\mathbf{q}} \sin^2(q_z d_{\parallel}) [J(0)S - \omega] g(\omega, \mathbf{q}). \quad (39)$$

Equations (38) and (39) resemble Eqs. (11) and (12) for a bond defect.

In the case of a vacancy, $J'S' = 0$, and we have, for $\Lambda^{(2)}(0)$,

$$\Lambda^{(2)}(0) = 1 - \frac{J(0)}{J_{\parallel}(0)} \frac{1}{N} \sum_{\mathbf{q}} \frac{\omega_{\parallel}^2(q_z)}{\omega^2(\mathbf{q})}, \quad (40)$$

which is of the order of $(J_{\perp}/J_{\parallel})^{1/2} \ll 1$.

If the defect is located in sublattice 2, then $T_2^{(2)}(\omega) = T_1^{(2)}(-\omega)$. The matrix elements of the full T matrix are

$$T_{11}^{(2)} = T_2^{(2)}, \quad T_{22}^{(2)} = T_1^{(2)}, \quad T_{12}^{(2)} = T_{21}^{(2)} = 0.$$

From Eqs. (36)–(40) we find that, in the case of a vacancy,

$$\begin{aligned} \text{Re}\Sigma^{(2)}(\omega, \mathbf{q}) &\approx -\omega_{\parallel}(q_z) \left[\frac{\Omega}{(2\pi)^3} \int \frac{d^3 q \omega_{q1}^2}{\omega_q^2} \right]^{-1} \\ &\approx -\omega_{\parallel}(q_z) [J_{\parallel}(0)/J_{\perp}(0)]^{1/2}. \end{aligned} \quad (41)$$

Thus, as in the case of the bond dilution, the renormalization of the spin-wave spectrum is small only at $x \ll x_0 \ll 1$.

The spin-wave damping

$$\Gamma^{(2)} = x \text{Im}\Sigma^{(2)}(\omega, \mathbf{q}) \approx x (J_{\parallel} J_{\perp}^2)^{-1} \omega^2(\mathbf{q}) \omega_{\parallel}^2(q_z) \propto q^4 \quad (42)$$

is smaller than the frequency ω_q if $x \ll x_0$.

When $J'S' \neq 0$, the function $\Lambda(0)$ is of the order of unity and the contribution of the representation A_{2U} to the self-energy is small at $x \ll 1$. The effect of the represen-

tation A_{1g} is more important in this case.

It follows from Eqs. (32) and (34) that the perturbation energy, corresponding to A_{1g} , is given by (the impurity is in the first sublattice)

$$V_1^{(1)} = S \frac{1}{N^2} \sum_{\mathbf{q}, \mathbf{q}'} [z_{\parallel} (c_{\mathbf{q}1}^{\dagger} + \gamma_{\parallel}(\mathbf{q})c_{-\mathbf{q}2}) (\varepsilon_{\parallel} c_{\mathbf{q}1} + \rho_{\parallel} \gamma_{\parallel}(\mathbf{q}') c_{-\mathbf{q}2}^{\dagger}) + z_{\perp} (c_{\mathbf{q}1}^{\dagger} + \gamma_{\perp}(\mathbf{q})c_{-\mathbf{q}2}) (\varepsilon_{\perp} c_{\mathbf{q}1} + \rho_{\perp} \gamma_{\perp}(\mathbf{q}') c_{-\mathbf{q}2})]. \quad (43)$$

Here

$$\begin{aligned} \gamma_{\parallel}(\mathbf{q}) &= 2^{-1} \sum_{\delta_{\parallel}} \exp(i\mathbf{q}\delta_{\parallel}) = \cos q_z d_{\parallel}, \\ \gamma_{\perp}(\mathbf{q}) &= 4^{-1} \sum_{\delta_{\perp}} \exp(i\mathbf{q}\delta_{\perp}) = 2^{-1} (\cos q_x d_{\perp} + \cos q_y d_{\perp}). \end{aligned} \quad (44)$$

Let us define two-component column and row vectors

$$|\eta^{\nu}(\mathbf{q})\rangle = \begin{bmatrix} 1 \\ \gamma_{\nu}(\mathbf{q}) \end{bmatrix}, \quad \langle \xi^{\nu}(\mathbf{q})| = S [\varepsilon_{\nu} z_{\nu}, z_{\nu} \rho_{\nu} \gamma_{\nu}(\mathbf{q})], \quad (45)$$

$\nu = \parallel, \perp$.

Then the solution of Eq. (9) with $V_{\mathbf{q}\mathbf{q}'}$ given by Eq. (43) can be written in the form

$$T_1^{(1)}(\mathbf{q}\mathbf{q}') = \frac{1}{\Lambda_1^{(1)}} [|\eta_{\mathbf{q}}^{\parallel}\rangle \langle \xi_{\mathbf{q}'}^{\parallel}| (1 - A_{\perp\perp}) + |\eta_{\mathbf{q}}^{\parallel}\rangle \langle \xi_{\mathbf{q}'}^{\perp}| A_{\parallel\perp} + |\eta_{\mathbf{q}}^{\perp}\rangle \langle \xi_{\mathbf{q}'}^{\parallel}| A_{\perp\parallel} + |\eta_{\mathbf{q}}^{\perp}\rangle \langle \xi_{\mathbf{q}'}^{\perp}| (1 - A_{\parallel\parallel})], \quad (46)$$

where

$$\begin{aligned} A_{\nu, \nu'} &= \frac{1}{N} \sum_{\mathbf{q}} \langle \xi^{\nu}(\mathbf{q}) | \hat{G}_0(\mathbf{q}) | \eta^{\nu'}(\mathbf{q}) \rangle \\ &= \frac{1}{N} \sum_{\mathbf{q}} z_{\nu} g(\mathbf{q}, \omega) \{ \varepsilon_{\nu} [\omega + J(0)S - J(\mathbf{q})S \gamma_{\nu}(\mathbf{q})] - \rho_{\nu} \gamma_{\nu}(\mathbf{q}) [\omega - J(0)S \gamma_{\nu}(\mathbf{q}) + J(\mathbf{q})S] \}, \end{aligned} \quad (47)$$

$$\Lambda_1^{(1)}(\omega) = (1 - A_{\parallel\parallel})(1 - A_{\perp\perp}) - A_{\perp\parallel} A_{\parallel\perp}. \quad (48)$$

The matrix $T_2^{(1)}$ for the antiferromagnet with the defect in the second sublattice is governed by Eq. (46) with ω replaced by $-\omega$ and the elements of the column $|\eta_{\mathbf{q}}^{\nu}\rangle$ (the row $\langle \xi_{\mathbf{q}}^{\nu}|$) interchanged.

A simple expression for $\Lambda(0)$ can be obtained when $J'_{\parallel}/J_{\parallel} = J'_{\perp}/J_{\perp} \equiv j$:

$$\Lambda_1^{(1)}(0) = j \left\{ 1 + J_{\parallel}(0)J_{\perp}(0)(j\beta - 1) \frac{1}{N} \sum_{\mathbf{q}} [\gamma_{\parallel}(\mathbf{q}) - \gamma_{\perp}(\mathbf{q})]^2 \omega^{-2}(\mathbf{q}) \right\}. \quad (49)$$

The function $\Lambda(0) = 0$ if $j = 0$, i.e., if the defect spin does not interact with the host spins. Clearly, this result does not depend on the value of the impurity spin.

In a quasi-1D antiferromagnet, the terms proportional to J_{\perp} can be neglected in all expressions, except $\omega(\mathbf{q})$. Then Eqs. (44), (45), (47), and (48) yield

$$\Lambda_1^{(1)}(\omega) = j [1 - \omega J_{\parallel}(0)(1 - \beta)g_0(\omega)] - \omega J_{\parallel}^{-1}(0) \{ 1 - j\beta + \omega g_0(\omega) [J_{\parallel}(0)(j - 1) + \omega(j\beta - 1)] \}, \quad (50)$$

where the local Green's function

$$g_0(\omega) = \frac{1}{N} \sum_{\mathbf{q}} [\omega^2 - \omega^2(\mathbf{q})]^{-1}. \quad (51)$$

The T matrix in this case can be written as

$$T^{(1)}(\mathbf{q}, \mathbf{q}') = \frac{2S}{\Lambda_1^{(1)}(\omega)} \begin{bmatrix} \varepsilon_{\parallel} & \rho_{\parallel} \gamma_{\parallel}(\mathbf{q}) \\ \varepsilon_{\parallel} \gamma_{\parallel}(\mathbf{q}) & \rho_{\parallel} \gamma_{\parallel}^2(\mathbf{q}) \end{bmatrix} + \frac{2S}{\Lambda_1^{(1)}(-\omega)} \begin{bmatrix} \rho_{\parallel} \gamma_{\parallel}^2(\mathbf{q}) & \varepsilon_{\parallel} \gamma_{\parallel}(\mathbf{q}) \\ \rho_{\parallel} \gamma_{\parallel}(\mathbf{q}) & \varepsilon_{\parallel} \end{bmatrix}. \quad (52)$$

It follows from Eqs. (52) and (36) that

$$\Sigma^{(1)}(\omega, \mathbf{q}) = \frac{S}{\omega(\mathbf{q})} \left\{ \varepsilon_{\parallel} S \left[\frac{1}{\Lambda_1^{(1)}(\omega)} + \frac{1}{\Lambda_1^{(1)}(-\omega)} \right] [J_{\parallel}(0) - J_{\parallel}(\mathbf{q})\gamma_{\parallel}(q_z)] + \omega [\varepsilon_{\parallel} - \rho_{\parallel} \gamma_{\parallel}^2(\mathbf{q})] \left[\frac{1}{\Lambda_1^{(1)}(\omega)} - \frac{1}{\Lambda_1^{(1)}(-\omega)} \right] \right\}. \quad (53)$$

In leading order in ω^2 and q^2 , we get

$$\begin{aligned} \Sigma^{(1)}(\omega, \mathbf{q}) = & (1-j^{-1})\omega_{\parallel}(q_z) \\ & + \omega(\mathbf{q})J_{\parallel}^2(0)(1-\beta)^2g_0(0) \end{aligned} \quad (54)$$

and

$$\begin{aligned} \text{Re}\Sigma^{(1)}(\omega, \mathbf{q}) \approx & -\omega(\mathbf{q})(1-\beta)^2 \frac{\Omega}{(2\pi)^3} \int d^3q \frac{J_{\parallel}^2(0)S^2}{\omega^2(\mathbf{q})} \\ \sim & -\omega(\mathbf{q})(\beta-1)^2 [J_{\parallel}(0)/J_{\perp}(0)]^{1/2}. \end{aligned} \quad (55)$$

We see that the renormalization of the spin-wave spectrum in quasi-1D antiferromagnets, doped by magnetic impurities, is enhanced if $S' \neq S$. The enhancement factor, as in antiferromagnets with vacancies, is equal to $[J_{\parallel}(0)/J_{\perp}(0)]^{1/2}$.

In the case of vacancies, the full renormalization is a sum of the term (55) with $\beta=0$ and the term given by Eq. (41):

$$\text{Rex}\Sigma(\omega, \mathbf{q}) = x x_0^{-1} [b_1 \omega(q_z) + b_2 \omega(\mathbf{q})], \quad (56)$$

where $b_1 \approx 1$, $b_2 \approx 1$.

For magnetic impurities with $S' \neq S$, only the contribution of the self-energy (55) is important. In this case the spin-wave damping

$$\Gamma^{(1)} = x \text{Im}\Sigma^{(1)} \approx x \frac{\omega^2(\mathbf{q})}{c_{0\parallel} c_{0\perp}^2} (\beta-1)^2 J_{\parallel}^2(0) \propto q^2. \quad (57)$$

The expression (57) determines the damping for all kinds of impurities we are considered here.

The relative damping

$$\frac{\Gamma^{(1)}}{\omega_q} \sim x \omega_q J_{\perp}^{-1} (\beta-1)^2 \quad (58)$$

is small for frequencies $\omega_q \ll T_N$ if $x \ll x_0$.

The unusual q^2 dependence of the damping stems from the local nonequivalence of the sublattices produced by the impurities with $S' \neq S$.¹¹

It should be noted that the representation A_{1g} is relevant also for the investigation of the impurity effect on the properties of quasi-2D magnets. However, for quasiplanar magnets the enhancement is smaller, of the order of $\ln(J_{\parallel}/J_{\perp})$.

Finally, we discuss the concentration dependence of the static uniform susceptibility χ . It appears that only the representation A_{1g} contributes to the renormalization of χ . The susceptibility increases as

$$\chi = (2J_{\parallel})^{-1} \left[1 + 2x(\beta-1)^2 \frac{\Omega}{(2\pi)^3} \int \frac{J_{\parallel}^2}{\omega^2(\mathbf{q})} d^3q \right]. \quad (59)$$

The renormalization, as for the spectrum, is of the order of x/x_0 for all kinds of defects we are considering.

IV. CONCENTRATIONS $x_0 \ll x \ll 1$: NONPERTURBATIVE TREATMENT

When $x \gg x_0$, the concentration expansion breaks down, though the dilution may be weak in the usual sense, $x \ll 1$. Thus a nonperturbative approach should

be used to study the concentration dependences of the spin-wave velocity and the susceptibility in the interval $x_0 \ll x \ll 1$.

We consider here only nonmagnetically diluted antiferromagnets. The interchain interaction will be treated in the molecular-field approach. This approximation is justified for the long-ranged $J_{\perp}(\mathbf{r})$ coupling. It is natural to suppose that, as usual, the molecular-field approach is good enough for the short-range interaction as well.

Using this approach, the energy per a segment containing n spins may be written as

$$\begin{aligned} E = & -J_{\parallel} S^2 \sum_{i,j=i\pm 1} \cos(\theta_i + \theta_j) \\ & - J_{\perp}(0) S^2 \sum_i \cos(\theta_i + \bar{\theta}) - HS \sum_i \sin\theta_i. \end{aligned} \quad (60)$$

Here θ_i is the canting angle of a spin \mathbf{S}_i in the external field \mathbf{H} and the angle $\bar{\theta}$ determines the mean value of the spin direction in a sublattice (Fig. 1). The sum in the first term is over the nearest neighbors in the chain.

Minimizing the energy at small H , when all θ_i are small, we obtain

$$\begin{aligned} \theta_1 + \theta_2 + h\theta_1 = \bar{H}, \quad \theta_n + \theta_{n-1} + h\theta_n = \bar{H}, \\ 2\theta_i + \theta_{i+1} + \theta_{i-1} + h\theta_i = \bar{H}, \quad i=2,3,\dots,n-1, \end{aligned} \quad (61)$$

where

$$h = J_{\perp}(0)/J_{\parallel}, \quad \bar{H} = H/J_{\parallel} - h\bar{\theta}. \quad (62)$$

Solving these equations, we find (see the Appendix) that for even segments ($n=2k$) all $\theta_i=0$, while for odd ones ($n=2k+1$)

$$\theta_1 = \theta_n = \bar{H}/hn. \quad (63)$$

Averaging with the nearest-neighbor distribution function

$$P(n) = x \exp(-nx),$$

we get that

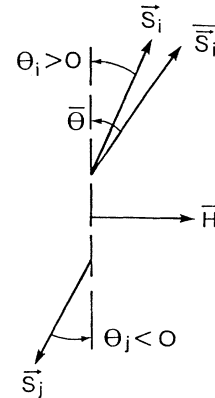


FIG. 1. Orientation of spins \mathbf{S}_{1i} (sublattice up) and \mathbf{S}_{2i} (sublattice down) which belong to the same segment. \mathbf{H} is the external magnetic field, and the angle $\bar{\theta}$ specifies the orientation of the sublattice magnetization.

$$\bar{\theta} = \frac{xH}{J_{\perp}(0)S} \ln x^{-1}. \quad (64)$$

To calculate χ , let us sum Eqs. (61). One gets

$$\sum_{i=1}^n \theta_i = \frac{1}{2}(\theta_1 + \theta_n) + \frac{1}{4}n\bar{H}.$$

Finally, averaging over n and using Eq. (63), we obtain the concentration dependence of the susceptibility per spin:

$$\chi = \frac{xS}{H} \sum_{i=1}^n \theta_i = J_{\perp}^{-1}(0)x^2 \ln x^{-1}. \quad (65)$$

The dependence $\chi(x)$ may be obtained qualitatively considering the orientation of the rigid spin segments in the external field. Segments with an even number of spins do not contribute to the low-field susceptibility, while the total spin of the odd segments is S , and there the contributions is $\chi_n \sim (nJ_{\perp})^{-1}$. Averaging χ_n leads to the dependence (65).

The deviation from the rigid segment picture is of the order of $x^{-1}(J_{\perp}/J_{\parallel})^{1/2} \ll 1$.

The spin-wave velocities can be determined from the Harris-Kirkpatrick relation¹¹

$$c_{\parallel, \perp} \propto (\sigma_{\parallel, \perp}/\chi)^{1/2}, \quad (66)$$

where $\sigma_{\parallel}, \sigma_{\perp}$ are the conductivities of a network with conductances J_{ij} between sites i and j .

According to Refs. 14 and 15, in a quasi-1D conductor $\sigma_{\parallel} \propto J_{\perp}x^{-2}$, $\sigma_{\perp} \propto J_{\perp}$. Thus Eq. (66) yields

$$\begin{aligned} c_{\parallel} &\propto J_{\perp}(0)x^{-2}(\ln x^{-1})^{-1/2}, \\ c_{\perp} &\propto J_{\perp}(0)x^{-1}(\ln x^{-1})^{-1/2}. \end{aligned} \quad (67)$$

When x increases, the ratio c_{\perp}/c_{\parallel} also increases; i.e., the spectrum becomes more isotropic.

V. CONCLUSIONS

We investigated the concentration dependences of the spectrum and transversal susceptibility. It was demonstrated that this dependence is strongly nonperturbative at small $x \ll 1$ concentrations. Although we studied mainly the simplest case of two-sublattice collinear spin ordering, our results are not restricted to a specific type of antiferromagnetic ordering. Only the strong spatial

anisotropy of the exchange coupling in the host material is important.

We considered above only the nearest-neighbor interactions in a chain which connects segments cut by vacancies. However, it is not difficult to take into account the interaction of the next-nearest neighbors as well. Calculating a new expression for $\Lambda(\omega)$, we have shown that this coupling J_1 may be neglected if $J_1/J_{\parallel} \ll (J_{\parallel}/J_{\perp})^{1/2}$. Thus, even if J_1 exceeds J_{\perp} , it may still be irrelevant.

If this inequality is not fulfilled, the segment coupling via the J_1 interaction is more important than that via the interchain coupling. Hence, at sufficiently high x , the susceptibility per segment with n spins is of the order of J_1^{-1} , and the mean value of χ per spin is $\propto xJ_1^{-1}$. This is in contrast with the dependence Eq. (65). Thus it is possible to find out experimentally whether the next-nearest-neighbor coupling or the interchain interaction is more important.

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APPENDIX

Let us introduce the new variables $\phi_i = \theta_i + \theta_{i+1}$ and sum any i th and $(i+1)$ th equations of the set (61). Then this set is rewritten in the form

$$\begin{aligned} (2+h)\phi_1 + \phi_2 &= 2\bar{H}, & (2+h)\phi_{n-1} + \phi_n &= 2\bar{H}, \\ (2+h)\phi_i + \phi_{i+1} + \phi_{i-1} &= 2\bar{H}, & i &= 2, 3, \dots, n-2. \end{aligned} \quad (A1)$$

The determinant D_{n-1} of the set (A1) may be calculated at $h=0$. It satisfies the recurrence relation

$$D_n = 2D_{n-1} - D_{n-2}. \quad (A2)$$

It follows from (A2) that $D_{n-1} = n$. Thus $\phi_1 = 2\bar{H}A_{n-1}n^{-1}$, where A_{n-1} differs from D_{n-1} by replacing any element in the first column by 1. Now it is easy to see that

$$A_{2k-1} = A_{2k} = k. \quad (A3)$$

Substituting the value of ϕ_1 into the first equation of the set (61), we arrive at the formula (63).

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