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Quantum spin liquid and antiferromagnetism

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A spin liquid concept for the Heisenberg Hamiltonian (spin $s = 1/2$) with antiferromagnetic
exchange interactions between nearest neighbors is developed. The spin liquid is described by the
Green's function method in the framework of a second-order theory. Equations are presented
for the self-consistent calculation of the parameters of the system and its thermodynamic
properties at all temperatures. A description of the spin system in the sc and bcc lattices is proposed
wherein it is treated as a spatially homogeneous spin liquid with a condensate and with a
singlet ground state. It is shown that the modulus of the "staggered" magnetization is expressed
uniquely in terms of a condensate at a boundary point of the Brillouin zone and is the long-
range order parameter. The existence region in temprature of the ordered state of the spin liquid
with a condensate ($T < T_0$) is wider than the existence region of the two-sublattice
antiferromagnetism ($T_0 < T_N$, where T_N is the Néel temperature), while the energy is lower. For
$T > T_0$ the system passes into an ordinary spin liquid state. © *2003 American Institute of
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1. MARSHALL EQUATION AND ANTIFERROMAGNETISM

The rigorous theoretical description of antiferromagnetic
(AF) ordering still remains an open problem after more than
half a century of history. The intensive discussion of the
necessary and sufficient conditions for the onset of long-
range AF order in a crystal has been going on for almost 50
years since a description was given in the framework of
mean (molecular) field theory and a quantum spin-wave
theory of antiferromagnetism was developed. The ground-
work for this problem was laid in Ref. 1.

The fundamental questions of the theory of antiferro-
magnetism are discussed, as a rule, on the basis of the iso-
tropic Heisenberg model ($s = 1/2$) with antiferromagnetic ex-
change interactions between nearest neighbors. The
Hamiltonian of the system

$$\mathcal{H} = \frac{1}{2} J \sum_{\mathbf{f}\Delta} \mathbf{s}_{\mathbf{f}} \cdot \mathbf{s}_{\mathbf{f}+\Delta}, \quad \mathbf{S} = \sum_{\mathbf{f}} \mathbf{s}_{\mathbf{f}} \tag{1}$$

is specified on an ideal lattice of dimensionality $d = 1,2,3$
with periodic boundary conditions: N is the number of sites,
$\mathbf{f}$ are the coordinates of the sites, $J > 0$ are the exchange
integrals between nearest neighbors, z is the number of near-
est neighbors, Δ are vectors connecting nearest neighbors,
$\mathbf{S}_{\mathbf{f}} = (s_{\mathbf{f}}^+, s_{\mathbf{f}}^-, s_{\mathbf{f}}^z)$ is the spin operator at site $\mathbf{f}$, and $\mathbf{S}$ is the total
spin operator. One of the main problems is to describe the
long-range antiferromagnetic order. The properties of the
system depend substantially on its dimensionality and geom-
etry.

According to the Merman–Wegner theorem (see Ref. 2),
long-range magnetic order in one- and two-dimensional sys-
tems (in the case of short-range exchange interactions) is
absent at any finite temperature $T \neq 0$. For a linear chain ($d
= 1$) there is an exact solution due to Hulthén:[2] the ground
state is a nondegenerate singlet ($S = 0$) with energy per bond

$\varepsilon_0 = 0.25 - \ln 2 = -0.4431$ (in units of the exchange). In a
square lattice ($d = 2$) it is only for $T = 0$ that a two-sublattice
AF state can be constructed and its energy ε_{AF} calculated in
the framework of the spin-wave theory. In a previous paper[3]
by the author it was shown on the basis of the spin liquid
concept that the ground state of a Hamiltonian spin system
on a square lattice is a singlet state with an energy
$\varepsilon_0 < \varepsilon_{AF}$, viz., $\varepsilon_0 = -0.352$.

Discussion of the problem of long-range AF order cen-
ters mainly on three-dimensional systems and consists in the
following.

On the one hand, there is the traditional approach to the
description of antiferromagnetism. In alternant lattices[1) (sc
and bcc and also the square lattice and the linear chain) a
"checkerboard" arrangement of spins is possible which can
be described by the Néel wave vector $|AF\rangle$ (an antiferro-
magnet with two mirror sublattices A and B). It is known
that such a function is an eigenfunction only for the operator
S^z with a total spin projection $S^z = S_A^z + S_B^z = 0$, but it is not
an eigenfunction for the Hamiltonian nor for the operator $\mathbf{S}^2$.
This means that the antiferromagnetic state is a state with an
indeterminate degeneracy.[4] However, the use of the approxi-
mate Néel function $|AF\rangle$, in which the sublattices are fixed
and long-range AF order is postulated, has its indisputable
advantages, since it permits calculation of the excitation
spectrum, the energy of the AF state with allowance for
transverse quantum spin fluctuations, the sublattice magneti-
zation $\bar{s}(T)$, and the Néel temperature T_N. The theory is
approximate and is valid only in the magnetically ordered
phase for $T \leq T_N$.

On the other hand, Marshall[5] (see Ref. 1) has stated a
theoremic assertion that the ground state of Hamiltonian (1)
with AF exchange interactions J between nearest neighbors
on alternant lattices is a nondegenerate singlet with $S = 0$. It

TABLE I. Main characteristics of a spin system with an antiferromagnetic exchange bond between nearest neighbors.

Lattice type	Singlet (SL and SLC)				AF		
	$\|\varepsilon_0\|$ Ref. 5	$\|\varepsilon_0\|$	$\|m_0\|$	τ_0	$\|\varepsilon_{AF}\|$	$\bar{s}(0)$	τ_N
Square	0.328	0.352	∞	–	0.335	0.3	0
sc	0.3007	0.312	0.434	0.213	0.296	0.432	0.163
bcc	0.2892	0.297	0.452	0.224	0.287	0.448	0.181

Note: $\varepsilon = \langle H \rangle / \langle zNJ/2 \rangle$ is the energy of the system at zero temperature (in units of the exchange per bond) in the singlet (0) and antiferromagnetic (AF) states; $|m_0|$ is the modulus of the "staggered" magnetization at $T=0$; $\bar{s}(0)$ is the AF order parameter (sublattice magnetization) at $T=0$; $\tau_0 = T_0/zJ$ is the temperature at which $|m|$ vanishes; $\tau_N = T_N/zJ$ is the Néel temperature.

has been *rigorously* proven only for the one-dimensional chain, while for $d=2$ and 3 dimensions there are enormous mathematical difficulties in constructing an exact singlet function, since the number of partial singlet functions contained in it and comprising the total set of states is factorially large in N. The absence of an exact singlet state makes it impossible to directly verify Marshall's assertion, which has a natural quantum mechanical basis. Indeed, Hamiltonian (1) commutes with any component of the total spin operator **S**, and its eigenfunctions are therefore eigenfunctions of the operators S^z and $\mathbf{S}^2$, while the eigenvalues $E(S)$ are classified according to the value of the total spin S ($0 \leq S \leq N/2$). According to Marshall, min $\{E(S)\} = E(0)$ corresponds to a singlet ground state with $S=0$. Marshall constructed an approximate singlet wave function and used a variational method to calculate the energy ε_0 in alternant lattices (see Table I). A comparison of the energies ε_0 (according to Marshall) and ε_{AF} shows that these energies are very close, but in the context of the approximations made (which are, generally speaking, different), it is impossible to conclusively decide the type of ground state.

The question arises: Can these two approaches be reconciled theoretically? Below, on the basis of the spin liquid concept developed by the author, a theoretical version of the description of the properties of the spin system in sc and bcc lattices is proposed which is valid at all temperatures. It should be noted that the spin liquid (SL) concept has a rather wide domain of applicability. In addition to two-dimensional systems, in the fcc lattice, because of frustration of the exchange J bonds and the presence of transverse quantum spin fluctuations, the AF state is absent at all temperatures, and the system is a spin liquid with a singlet ground state.[6] In this paper it is shown that in the sc and bcc lattices the ground state is a singlet (in accordance with Marshall), but for $T \leq T_0$ the system nevertheless has long-range order, which is described by the modulus of the "staggered magnetization." For $T > T_0$ the system is found in an ordinary SL state.

The results of the spin-wave theory and the main equations of the spin liquid theory are briefly set forth below in the framework of a unified Green's function method.[7]

2. SPIN-WAVE THEORY OF ANTIFERROMAGNETISM

On alternant lattices a "checkerboard" distribution is possible, with spins "up" (sublattice A, sites α) and "down"

(sublattice B, sites β); for nearest neighbors $\alpha + \Delta = \beta$, $\beta + \Delta' = \alpha'$. It is convenient to go over to a dimensionless Hamiltonian $h = H/zJ$; then all the energy parameters will be measured in units of zJ, including the temperature $\tau = T/zJ$.

The spin-wave theory is based on linearized first-order equations (the Tyablikov decoupling)

$$i\dot{s}^+_\alpha = \bar{s}\left(s^+_\alpha + \frac{1}{z} \sum_\Delta s^+_{\alpha+\Delta} \right), \quad i\dot{s}^+_\beta = -\bar{s}\left(s^+_\beta + \frac{1}{z} \sum_\Delta s^+_{\beta+\Delta} \right), \tag{2}$$

where $\langle s^z_\alpha \rangle = -\langle s^z_\beta \rangle \equiv \bar{s}$. After Fourier transformation with respect to the sublattices in the standard way, we find the Green's functions

$$\langle\langle S^+_A(\mathbf{q})|S^-_A(-\mathbf{q})\rangle\rangle_\omega = \frac{2\bar{s}(\omega + \bar{s})}{D(\mathbf{q},\omega)},$$

$$\langle\langle S^+_A(\mathbf{q})|S^-_B(-\mathbf{q})\rangle\rangle_\omega = \frac{2\bar{s}^2 \gamma_\mathbf{q}}{D(\mathbf{q},\omega)}, \tag{3}$$

where

$$D(\mathbf{q},\omega) = \omega^2 - \Omega^2_\mathbf{q}, \quad \Omega_\mathbf{q} = \bar{s}\sqrt{1 - \gamma^2_\mathbf{q}} \equiv \bar{s}\varepsilon_\mathbf{q},$$

$$\gamma_\mathbf{q} = \frac{1}{z}\sum_\Delta e^{i\mathbf{q}\cdot\Delta}. \tag{4}$$

We calculate the Fourier transforms of the correlation functions $\langle S^+_A(\mathbf{q})S^-_A(-\mathbf{q})\rangle$ and $\langle S^+_A(\mathbf{q})S^-_B(-\mathbf{q})\rangle$ according to the spectral theorem and then, using the sum rule $\langle s^+_\alpha s^-_\alpha \rangle = (1/2) + \bar{s}$, we obtain an equation for calculating the order parameter:

$$\bar{s}(\tau) = \frac{1/2}{I(\tau)}, \quad I(\tau) = \frac{1}{N}\sum_\mathbf{q} \frac{1}{\varepsilon_\mathbf{q}} \coth\left(\frac{\bar{s}(\tau)\varepsilon_\mathbf{q}}{2\tau} \right). \tag{5}$$

It follows from (5) that for $\tau = 0$

$$\bar{s}(0) = \frac{1/2}{I_1}, \quad I_1 = \frac{1}{N}\sum_\mathbf{q} \frac{1}{\varepsilon_\mathbf{q}}, \tag{6}$$

and for $\tau \to \tau_N$, $\bar{s} \to 0$, where $\tau_N = T_N/zJ$ is the Néel temperature, we have

$$\tau_N = \frac{1/4}{I_2}, \quad I_2 = \frac{1}{N}\sum_\mathbf{q} \frac{1}{\varepsilon^2_\mathbf{q}}. \tag{7}$$

The energy of the antiferromagnet (in units of J per bond) is equal to

$$\varepsilon_{AF}(\tau) = \frac{\langle H \rangle}{(1/2)zN \cdot J}$$

$$\approx -\left(\bar{s}^2(\tau) + \bar{s}(\tau)\frac{1}{N}\sum_\mathbf{q} \frac{\gamma^2_\mathbf{q}}{\varepsilon_\mathbf{q}} \coth \frac{\bar{s}(\tau)\varepsilon_\mathbf{q}}{2\tau} \right). \tag{8}$$

For three-dimensional alternant lattices we obtain the following numerical results (the sums over the first Brillouin zone are replaced by integrals with the densities of states given in the Appendix):

sc $(z=6)$: $\varepsilon_{AF}(0) = -0.297$, $\bar{s}(0) = 0.432$.

$\tau_N = 0.163$;

bcc $(z=8)$: $\varepsilon_{AF}(0) = -0.287$, $\bar{s}(0) = 0.448$. $\tag{9}$

$\tau_N = 0.181.$

3. QUANTUM SPIN LIQUID

Let us consider a spin system with the Hamiltonian (1) in a lattice of arbitrary dimensionality and geometry. We define a *spin liquid* as a system which is, on average, spatially uniform, with no breaking of the spin symmetry, and in which the spin correlation functions are isotropic,

$$\frac{1}{N}\sum_{\mathbf{f}} \langle s_{\mathbf{f}}^x s_{\mathbf{f+r}}^x\rangle = \frac{1}{N}\sum_{\mathbf{f}} \langle s_{\mathbf{f}}^y s_{\mathbf{f+r}}^y\rangle = \frac{1}{N}\sum_{\mathbf{f}} \langle s_{\mathbf{f}}^z s_{\mathbf{f+r}}^z\rangle \equiv \frac{1}{4} K_r \tag{10}$$

and depend only on the modulus of the distance $r = |\mathbf{r}|$, with $K_0 = 1$ (the sum rule); the average values for any component of a site spin and for any component of the total spin operator are equal to zero:

$$\langle s_{\mathbf{f}}^\alpha\rangle = 0, \quad \langle S^\alpha\rangle = 0, \quad \alpha = x,y,z \text{ or } +,-,z; \tag{11}$$

and the average value of a product of spin operators on an odd number of *different* sites is zero:

$$\langle s_{\mathbf{f}}^\alpha s_{\mathbf{m}}^\beta s_{\mathbf{n}}^\gamma\rangle = 0, \quad \mathbf{f} \neq \mathbf{m} \neq \mathbf{n}, \quad \dots. \tag{12}$$

Here and below the symbol $\langle \dots \rangle$ denotes a thermodynamic average at a temperature $\tau = T/zJ$ and over the ground state wave function for $\tau = 0$.

The whole set of properties of the spin liquid—the ground state, excitation spectrum, and thermodynamics—must be described on the basis of Hamiltonian (1) and postulates (10)–(12). It should be noted that postulate (12) is newly introduced by the author; its consequences will be demonstrated below. Later it will be shown that the ground state is the singlet state with total spin $S = 0$, which is equivalent to $\langle \mathbf{S}^2\rangle_{\tau=0} = 0$.

The properties of the SL state are determined mainly by the spatial and temperature dependence of the spin correlation functions $K_r(\tau)$. The energy of the SL state per bond in units of J is given by

$$\varepsilon = \frac{\langle H\rangle}{(1/2)_Z NJ} = -\frac{3}{4} K_1, \tag{13}$$

where $K_{|\Delta|} = -K_1$ ($K_1 > 0$) is the correlator between nearest neighbors. For description of the SL state we go over to the Fourier transforms of the spin operators and introduce the Fourier transform of the correlation function

$$K(\mathbf{q}) = \sum_{\mathbf{r}} e^{-i\mathbf{qr}} K_r = 4\langle s^z(\mathbf{q}) s^z(-\mathbf{q})\rangle$$

$$= 2\langle s^+(\mathbf{q}) s^-(-\mathbf{q})\rangle, \quad K_r = \frac{1}{N}\sum_{\mathbf{q}} e^{i\mathbf{q\cdot r}} K(\mathbf{q}) \tag{14}$$

with the obvious property $K(\mathbf{q}) = K(-\mathbf{q})$. To calculate $K(\mathbf{q})$ we use the method of two-time temperature Green's functions.[7] Because of the isotropicity of the correlators it is sufficient to calculate the retarded commutator Green's function

$$\langle\langle s^z(\mathbf{q})|s^2(-\mathbf{q})\rangle\rangle_\omega = G(\mathbf{q}, \omega).$$

where ω is a dimensionless spectral variable in terms of which $K(\mathbf{q})$ is found according to the spectral theorem.

Second-order equations and their linearization

The spin liquid theory is based on equations not lower than second order, since $\langle s_{\mathbf{r}}^\alpha\rangle = 0$ and the first-order equations cannot be linearized (as is done in spin-wave theory). The exact equations of motion have the form ($\hbar = 1$)

$$i\dot{s}_{\mathbf{f}}^+ = \frac{1}{z}\sum_{\Delta} (s_{\mathbf{f}}^z s_{\mathbf{f+\Delta}}^\tau - s_{\mathbf{f+\Delta}}^z s_{\mathbf{f}}^+),$$

$$i\dot{s}_{\mathbf{f}}^z = \frac{1}{2z}\sum_{\Delta} (s_{\mathbf{f}}^+ s_{\mathbf{f+\Delta}}^- - s_{\mathbf{f+\Delta}}^+ s_{\mathbf{f}}^-) \equiv M_{\mathbf{f}}, \tag{15}$$

$$i\dot{M}_{\mathbf{f}} = \frac{\partial^2 s_{\mathbf{f}}^z}{\partial t^2} = \frac{1}{2z^2}\sum_{\Delta} (s_{\mathbf{f}}^z - s_{\mathbf{f+\Delta}}^z) + R_{\mathbf{f}}, \tag{16}$$

where

$$R_{\mathbf{f}} = \frac{1}{z^2}\sum_{\Delta \neq \Delta'} [s_{\mathbf{f}}^z s_{\mathbf{f+\Delta}}^+ s_{\mathbf{f+\Delta'}}^- + (s_{\mathbf{f+\Delta-\Delta'}}^z - s_{\mathbf{f+\Delta'}}^z) s_{\mathbf{f}}^+ s_{\mathbf{f+\Delta}}^-$$

$$- s_{\mathbf{f+\Delta}}^z s_{\mathbf{f}}^+ s_{\mathbf{f+\Delta-\Delta'}}^-]. \tag{17}$$

Performing the linearization of the operator $R_{\mathbf{f}}$ according to the scheme

$$s_{\mathbf{f}}^z s_{\mathbf{n}}^+ s_{\mathbf{m}}^- \approx s_{\mathbf{f}}^z \alpha_{|\mathbf{n-m}|}\langle s_{\mathbf{n}}^+ s_{\mathbf{m}}^-\rangle = \frac{1}{2}\alpha_{|\mathbf{n-m}|} K_{|\mathbf{n-m}|} s_{\mathbf{f}}^z,$$

$$\mathbf{f} \neq \mathbf{n} \neq \mathbf{m}, \tag{18}$$

where $\alpha_{|\mathbf{n-m}|}$ are parameters which correct the decoupling, after Fourier transformation we obtain the Green's function of the linear second-order theory in the form

$$G(\mathbf{q}, \omega) = \frac{A_{\mathbf{q}}}{\omega^2 - \Omega_{\mathbf{q}}^2}, \quad A_{\mathbf{q}} = \frac{K_1}{2}(1 - \gamma_{\mathbf{q}}),$$

$$\gamma_{\mathbf{q}} = \frac{1}{z}\sum_{\Delta} e^{i\mathbf{q\cdot\Delta}}. \tag{19}$$

Here

$$\Omega_{\mathbf{q}}^2 = \lambda^2(1 - \gamma_{\mathbf{q}})(|\varepsilon_{\min}| + \gamma_{\mathbf{q}} + \delta), \quad \lambda^2 = \frac{\alpha_1 K_1}{2}, \tag{20}$$

where α_1 is a correction factor for the nearest neighbors, $\varepsilon_{\min}$ is the lower boundary of the spectrum γ_q, and the parameter δ is a complicated construction of correlators in the first, second, etc. coordination zones, multiplied by the corresponding correction factors. The parameter δ thereby reflects the effective correlations in the "expanded" cluster and will be calculated self-consistently later (its explicit form is unimportant). The spectral intensity of the Green's function (19) is equal to

$$J(\mathbf{q}, \omega; \tau) = \frac{e^{\omega/\tau}}{e^{\omega/\tau} - 1}\frac{A_{\mathbf{q}}}{2\Omega_{\mathbf{q}}}[\delta(\omega - \Omega_{\mathbf{q}}) - \delta(\omega + \Omega_{\mathbf{q}})],$$

$$\Omega_q \geq 0. \tag{21}$$

According to the spectral theorem we obtain for the single-time average

$$\langle s^z(\mathbf{q})s^z(-\mathbf{q})\rangle \equiv \frac{1}{4}K(\mathbf{q}) = \int_{-\infty}^{\infty} J(\mathbf{q},\omega;\tau)d\omega$$

$$= \frac{A_\mathbf{q}}{2\Omega_\mathbf{q}}\coth\left(\frac{\Omega_\mathbf{q}}{2\tau}\right)$$

or

$$K(\mathbf{q}) = \frac{K_1}{\lambda}\frac{1-\gamma_\mathbf{q}}{E_\mathbf{q}(\delta)}\coth\left(\frac{\lambda E_\mathbf{q}(\delta)}{2\tau}\right), \quad \Omega_q \equiv \lambda E_\mathbf{q}(\delta). \quad (22)$$

Expression (22) attests to the fact that the proposed version of the SL theory contains three unknown parameters which are functions of temperature: $K_1(\tau)$—the modulus of the correlator between nearest neighbors, $\lambda(\tau)$—the "stiffness" parameter of the excitation spectrum, and $\delta(\tau)$—the "pseudogap" in the spectrum. They must all be calculated self-consistently in accordance with three equations (see below); here $\delta = \delta(\tau) \geqslant 0$, as is necessary for the condition $\Omega_\mathbf{q} \geqslant 0$ or $E_\mathbf{q}(\delta) \geqslant 0$.

Self-consistency equations

Three parameters must be calculated self-consistently: K_1, λ, and δ. Using the definition (14) of the spatial correlators K_r, we obtain the system of equations

$$\begin{cases} K_0 = 1 = \dfrac{1}{N}\sum_\mathbf{q} K(\mathbf{q}) = \dfrac{K_1}{\lambda}I_0(\delta,\tau), \\[2mm] K_1 = \dfrac{1}{N}\sum_\mathbf{q}(-\gamma_\mathbf{q})K(\mathbf{q}) = \dfrac{K_1}{\lambda}I_1(\delta,\tau), \\[2mm] K_{\text{tot}} = \dfrac{1}{N}\sum_\mathbf{q}(\gamma_\mathbf{q})^2 K(\mathbf{q}) \\[2mm] \qquad = \dfrac{1}{z^2}\sum_{\Delta,\Delta'}K_{|\Delta+\Delta'|} = \dfrac{K_1}{\lambda}I_2(\delta,\tau), \end{cases} \quad (23)$$

where

$$I_n(\delta,\tau) = \frac{1}{N}\sum_\mathbf{q}(-\gamma_\mathbf{q})^n\frac{1-\gamma_\mathbf{q}}{E_\mathbf{q}(\delta)}\coth\left(\frac{\lambda E_\mathbf{q}(\delta)}{2\tau}\right),$$

$$n = 0,1,2. \quad (24)$$

From Eqs. (23) we obtain the formal solution (we omit the arguments of the functions)

$$\lambda = I_1, \quad K_1 = I_1/I_0, \quad K_{\text{tot}} = I_2/I_0, \quad \alpha_1 = 2I_0 I_1. \quad (25)$$

The equation for the parameter δ arises from the requirement of an exact value of the second moment[3,6,10] and has the form

$$M_2 = \frac{1}{8}\left(K_{\text{tot}} + \frac{K_1}{z}\right) = \frac{\lambda K_1}{4}P(\delta),$$

$$P(\delta) \equiv \frac{1}{N}\sum_\mathbf{q}(1-\gamma_\mathbf{q})E_\mathbf{q}\coth\left(\frac{\Omega_\mathbf{q}}{2\tau}\right). \quad (26)$$

Using solution (25), we obtain

$$P(\delta) = \frac{I_2(\delta) + I_1(\delta)/z}{2I_1^2(\delta)}, \quad \delta = \delta(\tau). \quad (27)$$

Thus the self-consistent second-order linear theory is based on satisfaction of the sum rule $K_0 = 1$, the definitions of the

correlators K_1 and K_{tot} [Eq. (23)], and the requirement of an exact value of the second moment, which leads to Eq. (27). This equation plays a fundamental role in the SL theory and its further generalizations.

The sums over the Brillouin zone for I_n and P are replaced by integrals with a density of states $D(\varepsilon)$ corresponding to a dispersion relation $\gamma_\mathbf{q} = \varepsilon$. An approximation for the density of states $D(\varepsilon)$ is given in the Appendix. Combining Eqs. (25) and (27), we obtain a system of three equations for the self-consistent calculation of the parameters of the SL:

$$\begin{cases} \lambda = I_1 \quad (a), \\[1mm] K_1 = I_1/l_0 \quad (b), \\[1mm] P = \dfrac{I_2 + l_1/z}{2I_1^2} \quad (c). \end{cases} \quad (28)$$

Here

$$l_n(\delta,t) = \int D(\varepsilon)(-\varepsilon)^n\frac{1-\varepsilon}{E(\varepsilon,\delta)}\coth\left(\frac{E(\varepsilon,\delta)}{2t}\right)d\varepsilon,$$

$$P(\delta,t) = \int D(\varepsilon)(1-\varepsilon)E(\varepsilon,\delta)\coth\left(\frac{E(\varepsilon,\delta)}{2t}\right)d\varepsilon.$$

$$E(\varepsilon,\delta) = \sqrt{(1-\varepsilon)(|\varepsilon_{\min}| + \varepsilon + \delta)}.$$

$$\varepsilon_{\min} \leqslant \varepsilon \leqslant 1, \quad t = \tau/\lambda. \quad (29)$$

We now show that the *ground state is a singlet* (total spin $S = 0$). We introduce the function (the average per spin of the square of the total spin of the system)

$$S^2(\tau) \equiv \frac{1}{N}\langle\mathbf{S}^2\rangle = \frac{1}{N}\sum_{\mathbf{fm}}\langle s_\mathbf{f}s_\mathbf{m}\rangle = \sum_\mathbf{r}\frac{1}{N}\sum_\mathbf{f}s_\mathbf{f}s_{\mathbf{f}_+\mathbf{r}}$$

$$= \frac{3}{4}\sum_\mathbf{r}K_\mathbf{r} = \frac{3}{4}K(0), \quad (30)$$

which is expressed in terms of the Fourier transform of the correlation function (22) for $\mathbf{q} = 0$. For $\tau \equiv 0$ it follows from (22) that $K(0) = 0$ and $S^2(0) = 0$, which proves that the ground state is a singlet. On the other hand, expression (30) can be considered as the limit

$$K(0) = \lim_{\mathbf{q}\to 0}K(\mathbf{q}) = \frac{K_1}{\lambda}\lim_{\mathbf{q}\to 0}\frac{1-\gamma_\mathbf{q}}{E_\mathbf{q}(\delta)}\coth\left(\frac{\lambda E_\mathbf{q}(\delta)}{2\tau}\right)$$

$$= \frac{4\tau}{\alpha_1(|\varepsilon_{\min}| + \gamma_0 + \delta)}. \quad (31)$$

Hence for $\tau \to 0$ we obtain $K(0) = 0$ (singlet) as before, but for $\tau \equiv 0$ triplet excitations arise in the system, and because of them $S^2(\tau) \neq 0$.

In Refs. 3 and 6 the thermodynamic properties of the SL in the square and fcc lattices were described at all temperatures on the basis of the theory set forth above and the self-consistency equations (28). It was shown there that the spatial correlation functions are sign-varying, with a magnitude that falls off with increasing distance, so that the SL contains short-range order of the antiferromagnetic type.

4. SPIN LIQUID WITH A "CONDENSATE" IN THE sc AND bcc LATTICES

Alternant lattices can be regarded as two interposed sublattices A (sites α) and B (sites β), independently of the existence of real magnetic sublattices. The Brillouin zone of these lattices contains a boundary point $\mathbf{Q}$ at which $\gamma_{\mathbf{Q}} = -1 = \varepsilon_{\min}$ and the spectrum $E_{\mathbf{Q}}(\delta) = \sqrt{2\,\delta}$. These points are $\mathbf{Q} = (\pi, \pi)$, $\mathbf{Q} = (\pi, \pi, \pi)$, and $\mathbf{Q} = (2\pi, 2\pi, 2\pi)$ for the square, sc, and bcc lattices, respectively. It follows from this that

$$\exp(i\mathbf{Q}\cdot\alpha) = 1, \quad \exp(i\mathbf{Q}\cdot\beta) = -1. \tag{32}$$

The function $S^2(\tau)$ was considered above [see Eq. (30)] and it was shown that $S^2(0) = 0$ (the ground state is a singlet) at $\tau = 0$; this is equivalent to the equation

$$K(0) = \sum_{\mathbf{r}} K_{\mathbf{r}} = 0 \quad (\tau = 0). \tag{33}$$

Taking relations (32) into account, we can write $K(\mathbf{Q})$ in the form

$$K(\mathbf{Q}) = \sum_{\mathbf{r}} e^{-i\mathbf{Q}\mathbf{r}} K_{\mathbf{r}} = \sum_{\mathbf{r}} |K_{\mathbf{r}}|. \tag{34}$$

Since $K_{\mathbf{r}}$ is sign-varying, in Eq. (33) there is complete compensation of all the terms in the sum, while the value of $K(\mathbf{Q})$ in (34) can be macroscopically large and proportional to the volume of the system ($\propto N$).

"Condensate" in the spin liquid

Under this assumption we write $K_{\mathbf{r}}$ in the form

$$K_{\mathbf{r}} = \frac{1}{N} \sum_{\substack{\mathbf{q} \\ (\mathbf{q} \neq \mathbf{Q})}} e^{i\mathbf{q}\cdot\mathbf{r}} K(\mathbf{q}) + e^{i\mathbf{Q}\cdot\mathbf{r}} \frac{K(\mathbf{Q})}{N},$$

$$\frac{K(\mathbf{Q})}{N} \equiv \frac{K_1}{\lambda} C. \tag{35}$$

where C is a "condensate" (a function of temperature) which is unknown *a priori*. Then

$$K_0 = 1 = \frac{1}{N} \sum_{\substack{\mathbf{q} \\ (\mathbf{q} \neq \mathbf{Q})}} K(\mathbf{q}) + \frac{K_1}{\lambda}C,$$

$$K_1 = \frac{1}{N} \sum_{\substack{\mathbf{q} \\ (\mathbf{q} \neq \mathbf{Q})}} (-\gamma_{\mathbf{q}}) K(\mathbf{q}) + \frac{K_1}{\lambda}C, \tag{36}$$

$$K_{\text{tot}} = \frac{1}{N} \sum_{\substack{\mathbf{q} \\ (\mathbf{q} \neq \mathbf{Q})}} \gamma_{\mathbf{q}}^2 K(\mathbf{q}) + \frac{K_1}{\lambda}C.$$

In relations (36) we go from summation to integration with a density of states $D(\varepsilon)$. We take into account the following circumstances. First, the lower limit of integration (because of the restriction $\mathbf{q} \neq \mathbf{Q}$) is equal to $-1 + \zeta$, where ζ is an infinitesimal quantity ($\zeta \rightarrow +0$). In the sc and bcc lattices the density of states $D(\varepsilon)$ goes to zero in a square-root manner at the boundaries of the spectrum. For this reason the value of ζ can be simply set equal to zero (this is the same situation as in the description of Bose condensation in a three-dimensional gas). Second, in the presence of the "condensate" C we assume $\delta = 0$ in the expression for $K(\mathbf{q})$. In that
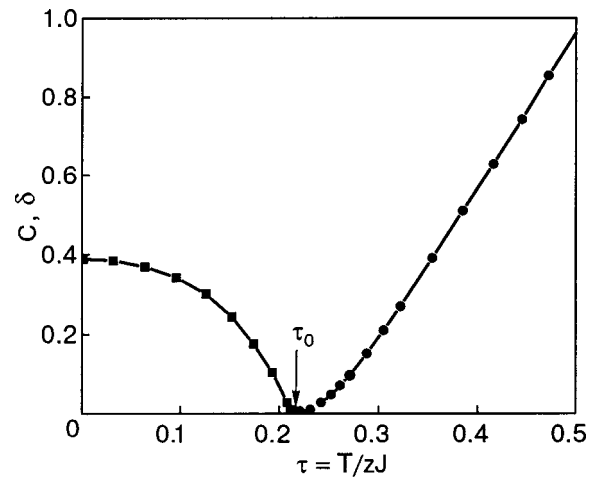

FIG. 1. Temperature behavior of the condensate (■) and the gap parameter (●) in the spin liquid in the sc lattice.

case the spectrum $E(\varepsilon, 0) = \sqrt{1 - \varepsilon^2}$ becomes symmetric with respect to inversion $\varepsilon \leftrightarrow -\varepsilon$, and the density of states has the same property: $D(\varepsilon) = D(-\varepsilon)$. When these symmetry properties are taken into account, the integrals become

$$I_0(t) = \int_{-1}^{1} D(\varepsilon) \frac{1}{\sqrt{1-\varepsilon^2}} \coth\left(\frac{\sqrt{1-\varepsilon^2}}{2t}\right) d\varepsilon.$$

$$I_1(t) = I_2(t) = \int_{-1}^{1} D(\varepsilon) \frac{\varepsilon^2}{\sqrt{1-\varepsilon^2}} \coth\left(\frac{\sqrt{1-\varepsilon^2}}{2t}\right) d\varepsilon,$$

$$P(t) = \int_{-1}^{1} D(\varepsilon) \sqrt{1-\varepsilon^2} \coth\left(\frac{\sqrt{1-\varepsilon^2}}{2t}\right) d\varepsilon \tag{37}$$

(the function $P(t)$ does not contain a "condensate" term).

Now relations (36) are written in the form

$$1 = \frac{K_1}{\lambda}(I_0 + C), \quad K_1 = K_{\text{tot}} = \frac{K_1}{\lambda}(I_1 + C), \tag{38}$$

and the self-consistency equation (27) assumes the simple form

$$P(I_1 + C) = \frac{z+1}{2z}. \tag{39}$$

From the numerical solution of the system of equations (38)–(39) at $\tau = 0$ we obtain

$$\lambda = 0.645, \quad K_1 = 0.416, \quad \varepsilon_0 = -0.312, \quad C = 0.389 \quad (\text{sc}).$$

$$\lambda = 0.607, \quad K_1 = 0.396, \quad \varepsilon_0 = -0.297,$$

$$C = 0.4185 \quad (\text{bcc}). \tag{40}$$

It follows from the results (40) that the energy of the singlet state in the presence of the condensate is lower than the energy of the AF state (see Table I). This proves Marshall's assertion that the ground state of the spin system on alternant lattices is a singlet.

Figure 1 shows the result of a calculation of the temperature behavior of the condensate in the sc lattice. This function goes to zero at a temperature $\tau_0 \approx 0.213$, which is higher than the Néel temperature $\tau_N = 0.163$ of the sc lattice. Thus the temperature region in which the spin liquid with the con-
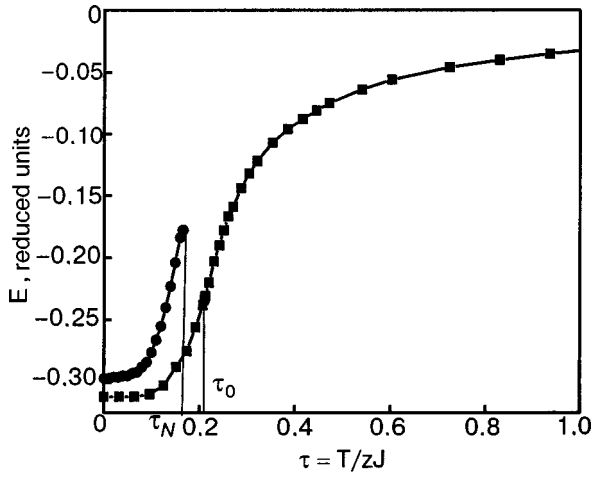
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FIG. 2. Energy of the antiferromagnetic state (●) calculated according to spin-wave theory for $\tau \leqslant \tau_N$, and the energy of the spin liquid in the sc lattice (■).

densate exists is wider than the existence region of the AF state, and the energy in this state $\varepsilon_{SLC} < \varepsilon_{AF}$. For $\tau > \tau_0$ a pseudogap δ begins to be "seeded" in the spectrum. To calculate it we use the same universal self-consistency equation (28c). The result is presented in Fig. 1. There is actually a phase transition

$$\text{SLC}(\delta=0, \ C\neq 0) \Rightarrow \text{SL}(\delta\neq 0, \ C=0),$$

and the properties of the system can be described at all temperatures. Figures 2 and 3 show the temperature behavior of the main parameters of the system. For comparison, the energy of the AF state (according to spin-wave theory) is shown in Fig. 2; it has a finite value at the Néel point, but above it the spin-wave theory is inapplicable.
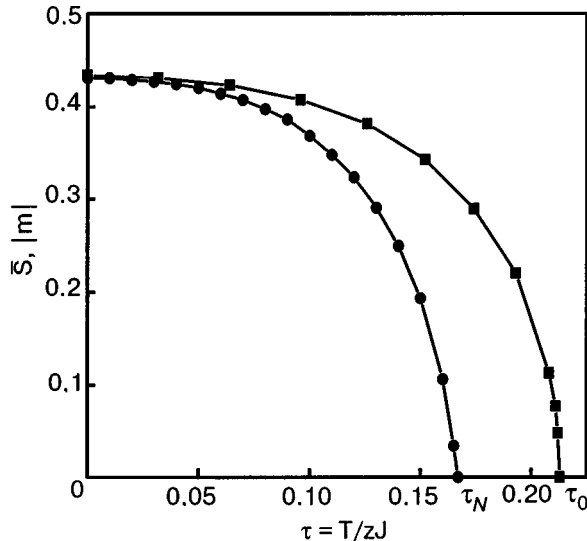

FIG. 3. Order parameters: $\bar{s}(\tau)$ is the relative magnetization of the sublattice in the AF state (●), and $|m(\tau)|$ is the modulus of the "staggered" magnetization in the spin liquid with the condensate (■) in the sc lattice.

"Staggered" magnetization, the "condensate," and long-range order

To explain the physical meaning of the "condensate," let us consider the square of the "staggered" magnetization (SM), which by definition[8] is equal to

$$m^2 = \left\langle \left(\frac{1}{N} \sum_{\mathbf{f}} e^{i\mathbf{Q}\cdot\mathbf{f}} \mathbf{s}_{\mathbf{f}} \right)^2 \right\rangle = \frac{1}{N} \sum_{\mathbf{r}} e^{i\mathbf{Q}\cdot\mathbf{r}} \frac{1}{N} \sum_{\mathbf{f}} \langle \mathbf{s}_{\mathbf{f}} \mathbf{s}_{\mathbf{f}+\mathbf{r}} \rangle$$

$$= \frac{1}{N} \sum_{\mathbf{r}} e^{i\mathbf{Q}\cdot\mathbf{r}} \frac{3}{4} K_{\mathbf{r}} = \frac{3}{4} \frac{K(\mathbf{Q})}{N} = \frac{3}{4} \frac{K_1}{\lambda} C, \tag{41}$$

where we have used the relation $2\mathbf{Q}\cdot\mathbf{f}=1$ and definition (35). It follows from (41) that the modulus of the SM is equal to

$$|m(\tau)| = \frac{\sqrt{3}}{2} \sqrt{\frac{K_1(\tau)}{\lambda(\tau)} C(\tau)},$$

or, when relations (38) are taken into account,

$$|m(\tau)| = \frac{\sqrt{3}}{2} \sqrt{\frac{C(\tau)}{I_0(\tau) + C(\tau)}}. \tag{42}$$

This function is compared with the order parameter $\bar{s}(\tau)$ in the AF state in Fig. 3; we note that

$$|m(0)| = 0.434, \quad \bar{s}(0) = 0.432 \quad \text{in sc},$$

$$|m(0)| = 0.452, \quad \bar{s}(0) = 0.448 \quad \text{in bcc}.$$

Thus the modulus of the "staggered" magnetization $|\mathbf{m}|$ is the order parameter in the quantum spin liquid for the sc and bcc lattices.[2)]

Long-range order

The presence of long-range order is detected from the behavior of the spatial correlation functions at $r \to \infty$ (the thermodynamic limit $N \to \infty$, $V \to \infty$, $N/V = \text{const}$ is understood; actually it is necessary to set $r \approx N^{1/d}$ equal to the maximum linear dimension of the system and then take the limit $N \to \infty$). For sign-varying correlation functions the long-range order is defined as a nonzero limit

$$C_\infty = \lim_{r \to \infty} |\langle \mathbf{s}_{\mathbf{f}} \mathbf{s}_{\mathbf{f}+\mathbf{r}} \rangle| = \frac{3}{4} \lim_{r \to \infty} |K_r| = \frac{3}{4} |K_\infty|. \tag{43}$$

Thus it is necessary to calculate K_r according to formula (35) for large values of r. In the sum (integral) over $\mathbf{q}$, as before, the main contribution comes from the vicinity of the point $\mathbf{q} = \mathbf{Q}$ (although the point $\mathbf{Q}$ itself is excluded). We set $\mathbf{q} = \mathbf{Q} + \mathbf{p}$ and perform the expansion $\gamma_{\mathbf{p}+\mathbf{Q}} = -\gamma_{\mathbf{p}} \approx -1 + p^2/z$; in addition, we formally drop the parameter δ in the spectrum. Then

$$K_\tau \approx e^{i\mathbf{Q}\cdot\mathbf{r}} \frac{K_1}{\lambda} \left[\frac{1}{(2\pi)^3} \int d^3\mathbf{p} e^{i\mathbf{p}\cdot\mathbf{r}} \frac{\sqrt{2z}}{\sqrt{p^2 + \kappa^2}} \right.$$

$$\left. \times \coth\left(\sqrt{\frac{2}{z}} \frac{\sqrt{p^2 + \kappa^2}}{2t} \right) + C \right], \tag{44}$$

where $\kappa^2 = z\delta$ and $t = \tau/\lambda$ (we note that the correlation length $\xi = 1/\kappa$). In the approximation $\coth x \approx 1/x$ the integral in (44) reduces to the form

$$2tz\frac{1}{(2\pi)^3}\int d^3\mathbf{p}\,\frac{e^{i\mathbf{p}\cdot\mathbf{r}}}{p^2+\kappa^2}\approx 2tz\frac{1}{4\pi}\frac{\exp(-\kappa r)}{r},\qquad(45)$$

which is well known in the Ornstein–Zernike theory. It follows from (45) that this integral goes to zero at large distances (even in the case $\delta=\kappa=0$), so that the long-range order is due to the "condensate" term, i.e.,

$$|K_\infty|=\frac{K_1}{\lambda}C\quad\text{or}\quad C_\infty=\frac{3}{4}|K_\infty|=m^2.\qquad(46)$$

Neutron scattering

In the general theory of inelastic neutron scattering (see, e.g., Ref. 7) the expression for the differential cross section for scattering contains the function

$$\Lambda(\mathbf{r},t)=\sum_{\alpha\beta}(\delta_{\alpha\beta}-e_\alpha e_\beta)\langle s_\mathbf{f}^\alpha(0)s_{\mathbf{f}+\mathbf{r}}^\beta(t)\rangle,\qquad(47)$$

where $\mathbf{e}=\mathbf{q}/q$, with $\mathbf{q}$ the neutron scattering vector. In a magnetically ordered state (F or AF), using the principle of decay of correlations at large distances, one performs a decoupling of the correlation function:

$$\langle s_\mathbf{f}^\alpha s_{\mathbf{f}+\mathbf{r}}^\beta\rangle\approx\langle s_\mathbf{f}^\alpha\rangle\langle s_{\mathbf{f}+\mathbf{r}}^\beta\rangle\propto\langle s_\mathbf{f}^z\rangle^2=\bar{s}^2,$$

which reduces to the square of the order parameter. In the spin liquid $\langle s_\mathbf{r}^\alpha\rangle=0$, and because of the isotropicity of the correlation functions (10), expression (47) assumes the form

$$\Lambda_{SL}(\mathbf{r},t)=\frac{1}{2}K_\mathbf{r}(t),\qquad(48)$$

i.e., it is expressed in terms of a time-dependent spatial correlation function.

Thus inelastic neutron scattering experiments measure the correlation function. The interpretation of the experimental data requires a separate and careful analysis.

5. SUMMARY

In this paper a theoretical version of the description of a spin system with an isotropic Heisenberg Hamiltonian (spin $s=1/2$, and an antiferromagnetic exchange only between nearest neighbors) as a spin liquid with a singlet ground state was proposed. It was shown that in three-dimensional alternant lattices (sc and bcc) at the boundary of the Brillouin zone there exists a "condensate" of excitations which determines the presence of a long-range order close to antiferromagnetic in the system. The two states (singlet and AF) are very similar (as was noted previously by Anderson[9]).

First, the excitation spectra are identical:

$$(\Omega_\mathbf{q})_{AF}=\bar{s}\sqrt{1-\gamma_\mathbf{q}^2},\quad(\Omega_\mathbf{q})_{SL}=\lambda\sqrt{1-\gamma_\mathbf{q}^2}\quad\text{at}\quad\delta=0,$$

where $\bar{s}$ is the order parameter (the relative sublattice magnetization) in the AF state, and $\lambda=\sqrt{\alpha_1 K_1/2}$, and K_1 is the modulus of the spin correlator between nearest neighbors in the SL.

Second, the spatial correlation functions of the spins are sign-varying.

Thus in the framework of the proposed version of the theory one is able to confirm Marshall's assertion that the

ground state in alternant lattices is a singlet, while preserving the presence of long-range magnetic order of the AF type.

The main results of this paper are as follows:

1. A definition of a spin liquid was given in the form of expressions (10)–(12) for an isotropic Heisenberg Hamiltonian. Postulate (12) was introduced here for the first time; it plays an important role in the construction of the self-consistent and internally closed theory of the SL.

2. The SL is described in the framework of a second-order theory by the Green's function method. In comparison with spin-wave theory (which contains only one order parameter ($\bar{s}(T)$) which is unknown *a priori*), in the SL theory there are three parameters: λ—the "stiffness" of the excitation spectrum, K_1—the modulus of the spin correlator in the first coordination zone (between nearest neighbors), and δ—the pseudogap in the spectrum (they are all functions of temperature).

3. A system of equations (37) was proposed for the self-consistent calculation of these parameters. As a result of the solution of this system (numerical and partly analytical) one can describe the thermodynamics of the SL at all temperatures.[3,6,10]

4. It was proved that the ground state of the SL is a singlet state.

5. A theoretical version of the description of a spin system in sc and bcc lattices as a spin liquid with a condensate (SLC) was proposed. The theory leads to the following results:

— the ground state is a singlet (total spin $S=0$, which corresponds to the rigorous quantum mechanical classification of states), the energy of the singlet state is lower than the energy of the AF state calculated according to spin-wave theory;

— the temperature dependence of the condensate was found; it vanishes at a critical temperature τ_0;

— it was shown that the modulus of the "staggered" magnetization $|m(\tau)|$ is expressed in terms of the condensate $C(\tau)$ and is the order parameter in the SLC; the existence region of the ordered state of the SL of the condensate is wider than that of the two-sublattice AF state, since $\tau_0>\tau_N$.

Thus the spin system has been described at all temperatures in the framework of the proposed theory.

APPENDIX

Approximation of the density of states for the dispersion relation $\gamma_\mathbf{q}=1/z\sum_\Delta e^{i\mathbf{q}\cdot\Delta}$, where Δ are the vectors connecting the nearest neighbors (the isoenergy surface $x=\gamma_\mathbf{q}$; the lattice parameter $a=1$).

Linear chain ($d=1$, $z=2$):

$$D(x)=\frac{1}{\pi}\frac{1}{\sqrt{1-x^2}},\quad|x|\leq 1.$$

The square lattice ($d=2$, $z=4$):

$$\gamma_\mathbf{q}=\frac{1}{2}(\cos q_x+\cos q_y),$$

$$D(x) = \frac{1}{\pi} - \left(\frac{1}{2} - \frac{1}{\pi}\right) \ln|x|, \quad |x| \leqslant 1.$$

The sc lattice ($d=3$, $z=6$):

$$\gamma_{\mathbf{q}} = \frac{1}{3}(\cos q_x + \cos q_y + \cos q_z),$$

$$D(x) = \begin{cases} 0.876, & |x| \leqslant 0.329, \\ 0.279 \dfrac{\sqrt{1-x^2}}{(x^2 - 0.09)^{0.3} + 10^8}, & 0.329 \leqslant |x| \leqslant 1. \end{cases}$$

The bcc lattice ($d=3$, $z=8$):

$$\gamma_{\mathbf{q}} = \cos\left(\frac{q_x}{2}\right)\cos\left(\frac{q_y}{2}\right)\cos\left(\frac{q_z}{2}\right),$$

$$D(x) = 0.431 \frac{(-\ln|x|)}{1 + 3x^2} + 0.186\sqrt{1-x^2}, \quad |x| \leqslant 1.$$

The fcc lattice ($d=3$, $z=12$):

$$\gamma_{\mathbf{q}} = \frac{1}{3}(c_x c_y + c_x c_z + c_y c_z), \quad c_j \equiv \cos\left(\frac{q_j}{2}\right),$$

$$D(x) = \begin{cases} A(x), & \text{if } -\dfrac{1}{3} \leqslant x \leqslant 0, \\ B(x), & \text{if } 0 \leqslant x \leqslant 1, \end{cases}$$

$$A(x) = -0.366664 \ln\left(0.0671182\left(x + \frac{1}{3}\right)\right) - 0.456693x,$$

$$B(x) = 0.226573\sqrt{1-x} + \frac{0.202745}{x + 0.151142} - 0.174703.$$

The density of states for the dispersion relation $\gamma_{\mathbf{q}}$ considered must satisfy the relations

$$\int D(x)dx = 1, \quad \int D(x)x\,dx = 0, \quad \int D(x)x^2\,dx = \frac{1}{z}.$$

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[1] Alternant lattices are those that can be represented in the form of two equivalent sublattices A and B interposed with each other in such a way that the nearest neighbors of sublattice A are sites of sublattice B and *vice versa*.

[2] In the antiferromagnetic state the "staggered" magnetization itself is the order parameter, $\mathbf{m}_{AF} = \langle 1/N \sum_{\mathbf{f}} e^{i\mathbf{Q}\cdot\mathbf{f}}\mathbf{S}_{\mathbf{f}}\rangle = \mathbf{e}1/N \sum_{\mathbf{f}} e^{i\mathbf{Q}\cdot\mathbf{f}}\langle S_{\mathbf{f}}^z\rangle = \mathbf{e}\bar{s}$ ($\mathbf{e}$ is the unit vector along the quantization axis). In the spin liquid $\mathbf{m}_{SL} = 0$, but $m_{SL}^2 \neq 0$, and $\sqrt{m_{SL}^2} \equiv |m|$ is the order parameter in the SLC.

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