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LOW-TEMPERATURE
SOLID-STATE PHYSICS

Magnetic-Field-Induced Phase Transition in a Two-Dimensional Quantum Magnet with Plaquette Distortion

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Abstract—Magnetic field effect on the structure of the ground state of a two-dimensional quantum Heisenberg magnet is analyzed. A plaquette representation is used to solve the self-consistent problem and calculate the collective excitation spectrum in a magnetic field. Conditions are found for quantum transition between nonmagnetic and oblique antiferromagnetic phases. The change in the ground state of the system is associated with disappearance of the gap in the spin excitation spectrum. Effects of frustration and magnetic field on the spectrum are analyzed. A phase diagram of stable singlet and magnetically ordered phases is presented.

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The introduction of a magnetic mechanism of the Cooper instability in high-temperature superconductors by Anderson [1] stimulated experimental and theoretical studies of quasi-low-dimensional magnets [2, 3]. Due to low dimension and bond frustration, quantum spin fluctuations in these materials can be sufficiently strong to change both the structure of the ground state and the low-temperature behavior of these systems. In particular, a sharp drop in magnetic susceptibility with decreasing temperature occurs in various quasi-two-dimensional magnetic systems, such as CaV_4O_9 [4], $Cu_3B_2O_6$ [5], $SrCu_4(BO_3)_2$ [6], and $(C_4H_{12}N_2)Cu_2Cl_6$ [7]. Analogous behavior is observed in quasi-one-dimensional systems undergoing the spin-Peierls transition [8], which consists in the low-temperature dimerization of an antiferromagnetic (AF) chain into a spin-singlet phase driven by magnetoelastic coupling.

In [9], the one-dimensional scenario of spin-singlet pairing was extended to two-dimensional systems. As AF long-range order breaks down in a square-lattice Heisenberg magnet with plaquette distortion (see [9, Fig. 1]), a gap opens in its excitation spectrum. The gap width depends on the interaction parameters of the model and external conditions, such as magnetic field strength. Experimental observations of magnetic-field-induced spin gap suppression and AF ordering in $TlCuCl_3$ crystals were reported in [10].

In this paper, the theoretical method used in [9] to analyze spin-singlet states of a two-dimensional magnet with plaquette distortion is generalized by constructing a plaquette representation that explicitly takes into account the sublattice tilting in a frustrated two-dimensional antiferromagnet at $H \neq 0$. The tilting is parameterized by an angle θ (as shown in Fig. 1), which can be found by exact treatment of the intraplaquette (strong) interactions. Thus, the magnetic order parameter is determined by the mean spin projections $\sigma_x = \langle S_1^x \rangle$ and $\sigma_z = \langle S_1^z \rangle$ at the first site (see site numbering in Fig. 1).

Magnetoelastic coupling is described by taking into account the dependence of exchange integrals on the intersite distance [9]. When an external magnetic field is applied, a modified self-consistent field is introduced as a vector with components parallel and perpendicular to the applied field, $\bar{H}_z = (2I^{ex} - (2J + J_1))\sigma_z$ and $\bar{H}_x = (2I^{ex} - (2J + J_1))\sigma_x$. The plaquette Hamiltonian is then written as

$$\begin{aligned} H_0 = & -2\bar{H}_x\sigma_x + \bar{H}_x D^x + 2\bar{H}_z\sigma_z - \bar{H}_z F^z \\ & + I^{in}(\mathbf{S}_1 \cdot \mathbf{S}_2 + \mathbf{S}_2 \cdot \mathbf{S}_3 + \mathbf{S}_3 \cdot \mathbf{S}_4 + \mathbf{S}_4 \cdot \mathbf{S}_1) \\ & + J^{in}(\mathbf{S}_1 \cdot \mathbf{S}_3 + \mathbf{S}_2 \cdot \mathbf{S}_4) \\ & - \mu g \mathbf{H} \cdot (\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4), \end{aligned} \quad (1)$$

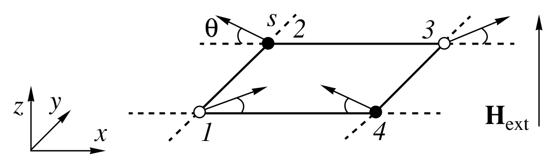

Fig. 1. Structure of the oblique phase in an external magnetic field.

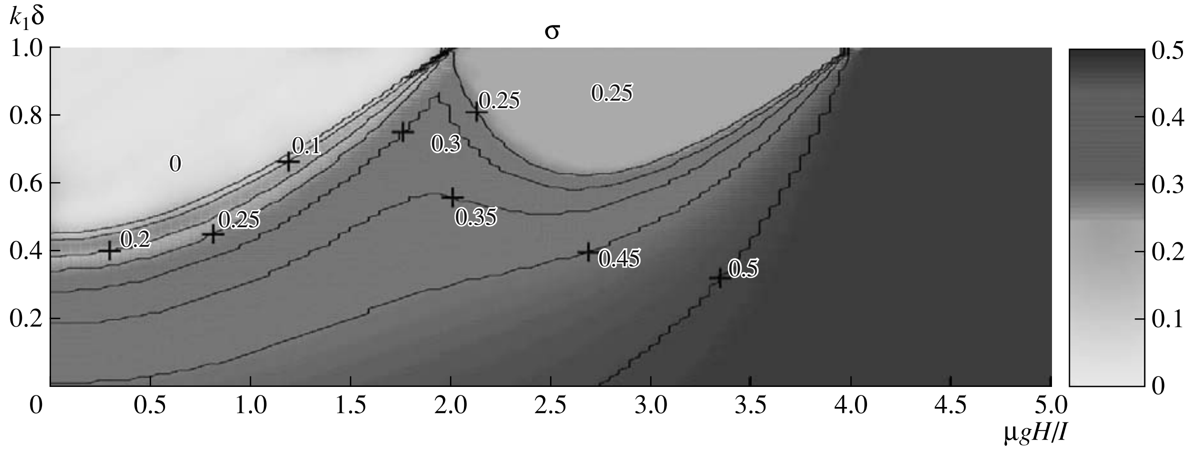

Fig. 2. Map of reduced magnetization as a function of magnetic field and distortion magnitude in the absence of frustration ($J = 0$).

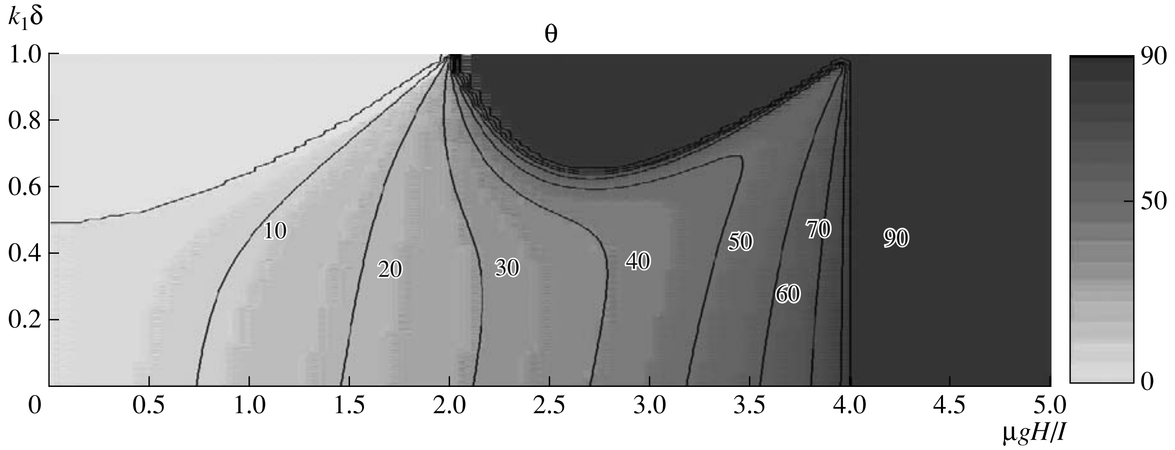

Fig. 3. Map of spin-tilt angle as a function of magnetic field and distortion magnitude in the absence of frustration ($J = 0$).

where

$$D^x = (S_1^x - S_2^x + S_3^x - S_4^x),$$
$$F^z = (S_1^z + S_2^z + S_3^z + S_4^z). \tag{2}$$

The x and z components of magnetization are found from the self-consistency conditions

$$\sigma_z = \frac{1}{4}\langle\psi_0(\sigma_x, \sigma_z)|S_1^z + S_2^z + S_3^z + S_4^z|\psi_0(\sigma_x, \sigma_z)\rangle,$$
$$\sigma_x = \frac{1}{4}\langle\psi_0(\sigma_x, \sigma_z)|S_1^x - S_2^x + S_3^x - S_4^x|\psi_0(\sigma_x, \sigma_z)\rangle. \tag{3}$$

The total magnetization and the spin-tilt angle are calculated as

$$\sigma = \sqrt{\sigma_x^2 + \sigma_z^2}, \quad \tan\theta = \sigma_z/\sigma_x. \tag{4}$$

The grayscale maps in Figs. 2 and 3 represent the total magnetization σ and the spin-tilt angle, respectively, as functions of distortion magnitude and applied field strength in the absence of frustration. White and black areas correspond to zero and maximum mean magnetizations, respectively (the latter being 1/2). Note that a collinear phase emerges as a superposition of four states in which three spins in a plaquette are parallel and the fourth one is antiparallel to the field. The resulting mean magnetization per site is 1/4. The collinearity of these states is obvious from Fig. 3. As expected, magnetic field restricts the region of stable spin-singlet states: a stronger distortion is required for transition to the nonmagnetic phase to occur (for $J = 0$, the transition point is $k_1\sigma \approx 0.42$). Conversely, a frustrated coupling J contributes to magnetic disorder and expansion of the spin-singlet region. A sufficiently strong magnetic field ($\mu gH \sim 4I$) drives the system into a ferromagnetic state.

Another trend observed at high distortion magnitudes is illustrated by Fig. 4, where the mean magnetization and its component σ_z are plotted versus field strength for $k_1\delta = 0.67$. The magnetization decreases as the field strength exceeds $2I$, whereas its component parallel to the field monotonically increases because of

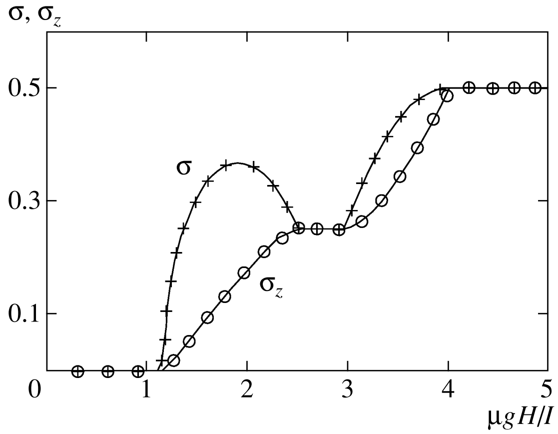

Fig. 4. Total magnetization and magnetization component parallel to magnetic field vs. field strength at $k_1\delta = 0.67$ ($J = 0$).

spin tilting. Note also that the magnetization curve has a plateau corresponding to the collinear phase.

After the physics of the ground state in various parameter regions has been elucidated, the elementary excitation spectrum can be found by a method analogous to that used in [9]. However, spin tilting makes calculations for $\mathbf{H} \neq 0$ more difficult to perform.

The numerical results presented in Fig. 5 illustrate the magnetic-field-induced change in the excitation spectrum of an unfrustrated system (with $J = 0$) at $k_1\delta = 0.5$. The distortion magnitude is chosen so that the system is disordered in zero magnetic field. The lower branch of the spectrum obtained for the spin-singlet phase splits into three in a weak field (see Fig. 5b), which leads to a narrower energy gap. At the point of transition to a magnetically ordered phase ($\mu gH = 0.5$), the gap closes in accordance with Goldstone's theorem (Fig. 5c). With further increase in field strength, the lower branch remains gapless while the other two move toward higher energies.

According to our calculations, the gap width is a linearly decreasing function of field strength when the distortion magnitude is held constant. Using the equation for the spin-singlet gap in zero field derived in [9], we can write an expression for the critical field as a function of distortion magnitude and degree of frustration:

$$\mu gH = I\left[\frac{11}{3}(k_1\delta)^2 + 2k_1\delta - \frac{5}{3} + 4\frac{J}{I}\left(1 + k_1\delta - \frac{1}{3}k_2\delta(1 + k_1\delta)\right)\right]^{1/2}. \quad (5)$$

The distortion energy per plaquette can be expressed as $E_{el} = 2\tilde{C}\delta^2 = 2Cd^2$, where C is an effective magne-

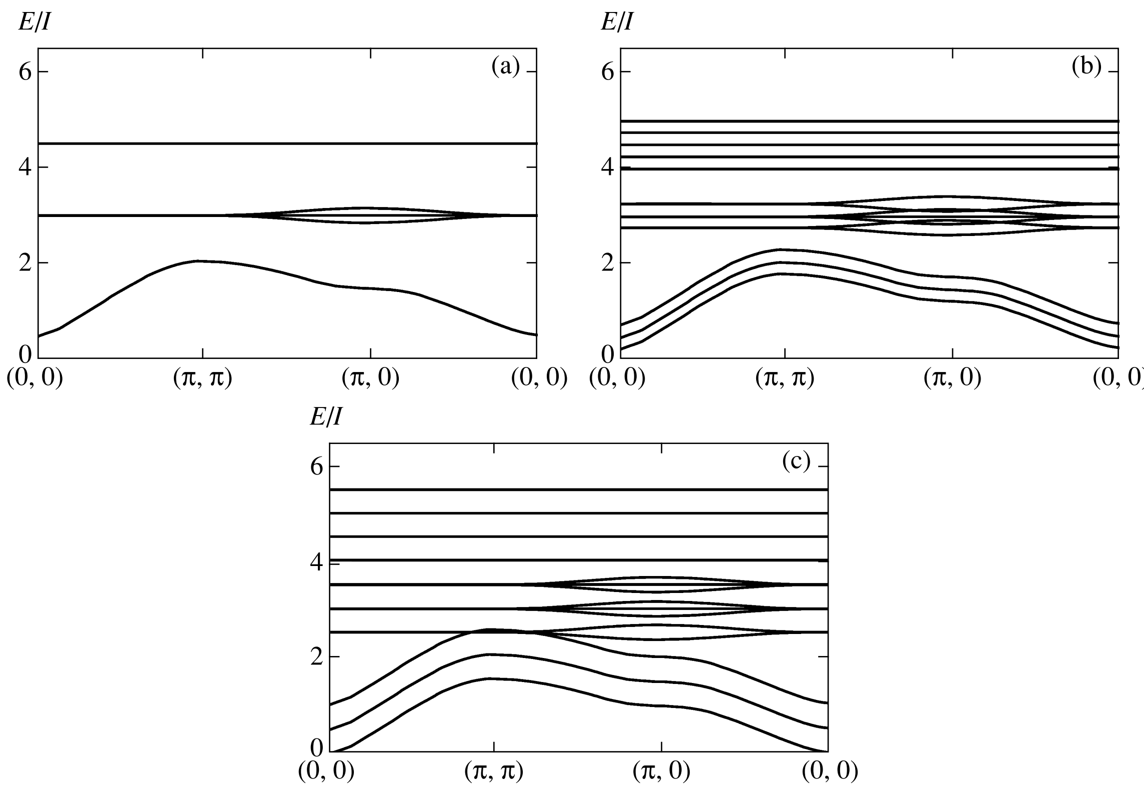

Fig. 5. Change in the elementary excitation spectrum for an unfrustrated system ($J = 0$) at $k_1\delta = 0.5$ and $H = 0$ (a), $0.25I$ (b), and $0.5I$ (c).

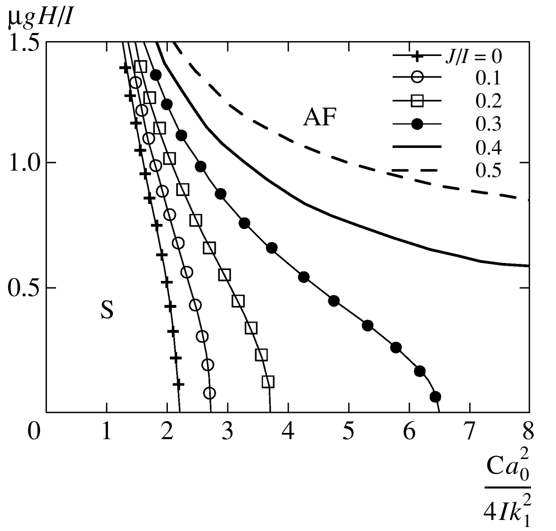

Fig. 6. Phase diagram of frustrated quantum two-dimensional quantum magnet with $k_2 = k_1$: S and AF correspond to spin-singlet and oblique antiferromagnetic phases, respectively.

toelastic coupling constant and d is strain. Then, the equilibrium value of distortion magnitude δ can be found by solving the equation

$$\frac{\partial E_m(\delta)}{\partial \delta} + 4\tilde{C}\delta = 0. \qquad (6)$$

At the point of transition to the spin-singlet phase, the value of δ must not only satisfy Eq. (6), but also correspond to the critical value of $(k_1\delta)_c$ corresponding to phase transition. Combining these two conditions, we obtain an equation that determines the boundary between antiferromagnetic and spin-singlet plaquette phases:

$$\frac{Ca_0^2}{4Ik_1^2} = \lambda\left(\frac{J}{I}, \frac{k_2}{k_1}, H\right), \qquad (7)$$

where

$$\lambda\left(J, \frac{k_2}{k_1}\right) = -\frac{1}{4(k_1\delta)_c}\left(\frac{\partial E_m}{\partial(k_1\delta)}\right)_{k_1\delta = (k_1\delta)_c}. \qquad (8)$$

Having found λ as a function of J/I, k_2/k_1, and field strength, we uniquely determine the phase diagram. Figure 6 shows the magnetic field that destroys the spin-singlet phase versus $Ca_0^2/4Ik_1^2$ for several values of degree of frustration.

When $J > 0.42I$, only the spin-singlet phase is stable in zero field for any value of λ. (The transition to the AF phase is left outside the scope of the present analysis, because it corresponds to different magnetic ordering.)

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