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2007 J. Phys.: Condens. Matter 19 156212

(http://iopscience.iop.org/0953-8984/19/15/156212)

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **19** (2007) 156212 (13pp) doi:10.1088/0953-8984/19/15/156212

Misalignment of on-site spins of localized and itinerant electrons in the double-exchange model with Coulomb repulsion

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Received 5 January 2007, in final form 9 February 2007
Published 21 March 2007
Online at stacks.iop.org/JPhysCM/19/156212

Abstract

We show that the on-site spins of localized and itinerant electrons in the double-exchange model are misaligned when the on-site Coulomb repulsion is large enough. To explore this phenomenon we use the Schwinger-boson representation of the localized spins and introduce two spin-singlet fermion operators. In terms of the new Fermi fields, the on-site Hund's interaction is in a diagonal form and the true magnons of the system are identified. The singlet fermions can be understood as electrons dressed by a cloud of repeatedly emitted and reabsorbed magnons. The quantum phase transition between ferromagnetism and the new phase is studied by varying the Coulomb repulsion for different values of parameters in the theory such as the Hund's coupling and chemical potential.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The spin-fermion model describes materials which get their magnetic properties from a system of localized magnetic moments being coupled to conducting electrons. The model is known as an s–d (or s–f) model, in which the electrons are separated into delocalized s electrons and localized d(f) electrons. The names of the s and d(f) electrons do not necessarily mean that the orbital electron states are of corresponding type. They are introduced to distinguish the localized from delocalized electrons. The model appears in the literature also as the ferromagnetic Kondo lattice model (FKLM) or the double-exchange model (DEM) [1–7].

The double-exchange model is a widely used model for manganites [1, 8]. In isolation, the ions of Mn have an active 3d shell with five degenerate levels. The degeneracy is presented due to rotational invariance within angular momentum $l = 2$ subspace. The crystal environment results in a particular splitting of the five d orbitals (*crystal field splitting*) into two groups: the e_g and t_{2g} states. The electrons from the e_g sector, which form a doublet, are removed upon hole

doping. The t_{2g} electrons, which form a triplet, are not affected by doping, and their population remains constant. The Hund rule enforces alignment of the three t_{2g} spins into a $S = 3/2$ state. Then, the t_{2g} sector can be replaced by a *localized spin* at each manganese ion, reducing the complexity of the original five-orbital model. The next drastic simplification is that only one e_g orbital is available at each site. To justify this one can assume [8] that a static Jahn–Teller distortion leads to a splitting of the degenerate e_g levels, allowing one to keep only one active orbital. The only important interaction between the two sectors is the Hund coupling between localized t_{2g} spins and mobile e_g electrons.

The double-exchange model has a rich phase diagram, exhibiting a variety of phases, with unusual ordering in the ground states. The procedures followed to obtain the phase diagram are different: numerical studies [9], dynamical mean field theory [10], and analytical calculations [11, 12]; but four phases have been systematically observed: (i) antiferromagnetism (AF) at a density of mobile electrons $n = 1$, (ii) ferromagnetism (FM), (iii) phase separation (PS) between FM and AF phases, and (iv) spin incommensurable (IC) phase. The competition between spin spiral incommensurate order or phase separation and canted ferromagnetism is also a topic of intensive study [11–13]. The phase diagram becomes more rich if the orbital degeneracy is accounted for [14].

The simplest but realistic Hamiltonian for the double-exchange model has the form

$$H = -t \sum_{\langle ij \rangle} \left(c_{i\sigma}^{\dagger} c_{j\sigma} + \text{h.c.} \right) - J_{\text{H}} \sum_{i} \mathbf{S}_i \cdot \mathbf{s}_i \tag{1}$$

where $c_{i\sigma}^{\dagger}$ and $c_{i\sigma}$ are creation and destruction operators for mobile electrons, $s_i^{\mu} = \frac{1}{2} \sum_{\sigma\sigma'} c_{i\sigma}^{\dagger} \tau_{\sigma\sigma'}^{\mu} c_{i\sigma'}$, with the Pauli matrices (τ^x, τ^y, τ^z), is the spin of the conduction electrons, and $\mathbf{S}_i$ is the spin of the localized electrons. The sums are over all sites of a three-dimensional cubic lattice, and $\langle i, j \rangle$ denotes the sum over the nearest neighbours. In equation (1) the hopping amplitude and the Hund coupling between localized and mobile electrons are positive.

The Hamiltonian (1) of the DEM is quadratic with respect to the fermions $(c_{i\sigma}^{\dagger}, c_{i\sigma})$. Averaging in the subspace of the itinerant electrons, one obtains an effective Heisenberg-like model in terms of core spins $\mathbf{S}_i$ [15]. In the small-J_{H} limit, Ruderman–Kittel–Kasuya–Yosida (RKKY) theory is recovered. The subtle point is that if we use a Holstein–Primakoff representation for the localized spins $\mathbf{S}_i$, the creation and annihilation Bose operators do not describe the true magnon of the system [16]. The true magnons are transversal fluctuations corresponding to the total magnetization which includes both the spins of localized and delocalized electrons. Therefore the RKKY validity condition requires not only small Hund's coupling, but it also requires the charge carrier density to be small, which in turn means that the magnetization of the mobile electrons is inessential.

An attempt to introduce the 'true magnon' is achieved in [17] and [18]. The effective Hamiltonians, and the bosonic (magnon) and fermionic sectors are constructed in $S^{-\frac{1}{2}}$ expansion up to the fourth order. Only the quadratic part in the Hund's term is kept. The four-fermion terms of order $\frac{1}{S}$ are dropped.

Since the only interaction between localized and delocalized electrons is the Hund coupling, it is desirable to treat the on-site term in the Hamiltonian (1) exactly. To realize this we introduce two spin-singlet Fermi fields. Originally they were introduced in [19] to explore the double-exchange model and in [20] in the context of the t–J model. In terms of the singlet Fermi fields the Hund's term is in a diagonal form, the spin variables are removed, and one can treat it exactly [21]. An analogous technique is used in [12].

A more realistic DEM would account for the on-site Coulomb repulsion. The Coulomb (Hubbard) interaction has a profound effect on the band structure, magnetic ground state (magnetic configurations), and transport properties of spin-fermion systems. The results of

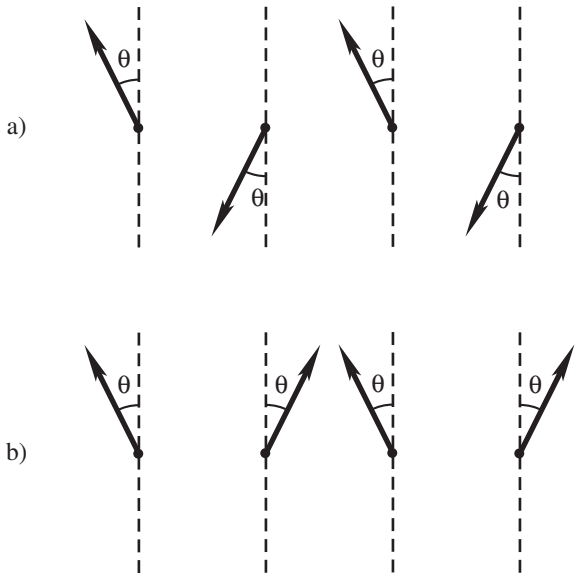

Figure 1. Sketch of two sublattice spin-canted states: (a) canted antiferromagnetism, (b) canted ferromagnetism.

the electron–electron repulsion depend on parameters in the theory such as doping, band width, and temperature. While some effects of the Hubbard term have been addressed in the past by means of mean-field theory [22–24], local spin-density approximation (LSDA) and LSDA + U calculations [25], and dynamical mean-field theory [26], its impact has not been fully appreciated so far.

In the present paper we study canted ferromagnetism in the double-exchange model with on-site Coulomb repulsion. Usually the canted magnetism is considered as a two-sublattice spin configuration with neighbouring lattice spins misaligned by an angle θ (figure 1). de Gennes first observed that in the double-exchange model a spin-canted state interpolates between ferromagnetic and antiferromagnetic order [27]. The canted phase is a part of the phase diagram of the mixed-spin $(S_1 > S_2)J_1 - J_2$ Heisenberg model on a square lattice [28]. Finally, a canted phase appears in the lattice models of quantum rotors [29].

In the present paper we show that *ferromagnetism with on-site spins of localized and delocalized electrons misaligned* (figure 2) emerges in the double-exchange model when the Coulomb repulsion is large enough. We study the quantum phase transition between ferromagnetic and on-site canted orders when the Coulomb repulsion is varied for different values of parameters in the theory such as the Hund's coupling and chemical potential.

The paper is organized as follows. In section 2 we study the double-exchange model (1) supplemented with an antiferromagnetic Heisenberg interaction between nearest-neighbour core spins. The Schwinger-boson representation of the localized spins is used and two spin-singlet fermion operators are introduced. In terms of the new Fermi fields the on-site Hund's interaction is in a diagonal form and the true magnons of the system are identified. The singlet fermions can be understood as electrons dressed by a cloud of repeatedly emitted and reabsorbed magnons. On integration over the singlet fermions, we obtain an effective Heisenberg-like theory. Positivity of the spin-stiffness, as a function of Hund's coupling J_{H} and charge carrier density, is the condition for stable ferromagnetism. The phase boundary is depicted in the case of both zero and non-zero antiferromagnetic exchange. Section 3 is

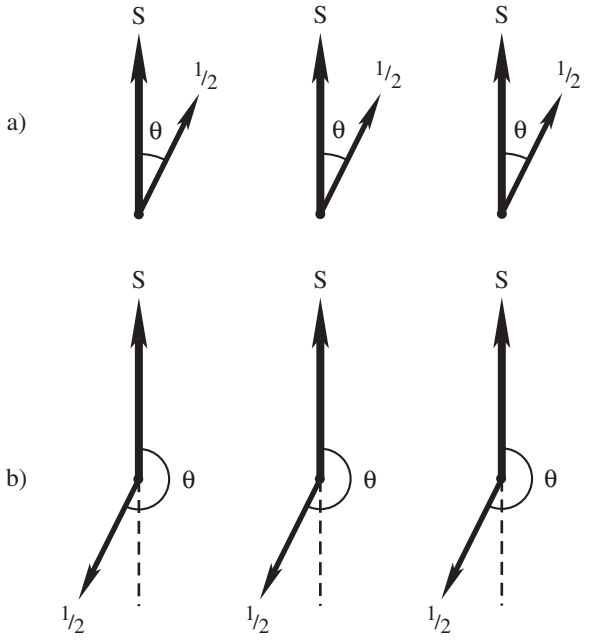

Figure 2. Sketch of on-site spin-canted states: (a) when $J_{\rm H} > 0$, (b) when $J_{\rm H} < 0$.

devoted to the on-site canted ferromagnetism in the DEM with on-site Coulomb repulsion. The theory rewritten in terms of Schwinger-bosons and spin-singlet fermions is a $U(1)$ gauge-invariant theory. We show that the on-site canted state is a state with spontaneously broken gauge symmetry. The quantum phase transition between ferromagnetism and the canted phase is studied by varying the parameters in the theory. A summary in section 4 concludes the paper.

2. Magnons in the double-exchange model

We consider a theory with the Hamiltonian

$$h = H - \mu N = -t \sum_{\langle ij \rangle} \left(c^{\dagger}_{i\sigma} c_{j\sigma} + \text{h.c.} \right) - \mu \sum_i n_i + J_{\rm AF} \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - J_{\rm H} \sum_i \mathbf{S}_i \cdot \mathbf{s}_i \qquad (2)$$

where μ is the chemical potential, and $n_i = c^{\dagger}_{i\sigma} c_{i\sigma}$. The antiferromagnetic Heisenberg term ($J_{\rm AF} > 0$) is very important for the manganites. In the limit when all e_g electrons are removed, and the system is without mobile electrons, the t_{2g} electrons induce an antiferromagnetic Heisenberg exchange between nearest neighbours, leading to standard antiferromagnetism. The most prominent example is $CaMnO_3$ [8].

In terms of Schwinger-bosons $(\varphi_{i,\sigma}, \varphi^{\dagger}_{i,\sigma})$ the spin operators have the following representation:

$$\vec{S}_i = \tfrac{1}{2} \varphi^{\dagger}_{i\sigma} \vec{\tau}_{\sigma\sigma'} \varphi_{i\sigma'}, \qquad \varphi^{\dagger}_{i\sigma} \varphi_{i\sigma} = 2S. \qquad (3)$$

The partition function can be written as a path integral over the complex functions of the Matsubara time $\varphi_{i\sigma}(\tau)$ $(\varphi^{\dagger}_{i\sigma}(\tau))$ and Grassmann functions $c_{i\sigma}(\tau)$ $(c^{\dagger}_{i\sigma}(\tau))$.

$$\mathcal{Z}(\beta) = \int \mathrm{d}\mu \left(\varphi^{\dagger}, \varphi, c^{\dagger}, c \right) \mathrm{e}^{-A} \qquad (4)$$

with an action given by the expression

$$A = \int_0^\beta \mathrm{d}\tau \left[\sum_i \left(\varphi_{i\sigma}^\dagger(\tau)\dot{\varphi}_{i\sigma}(\tau) + c_i^\dagger(\tau)\dot{c}_i(\tau) \right) + h(\varphi^\dagger, \varphi, c^\dagger, c) \right], \tag{5}$$

where β is the inverse temperature and the Hamiltonian is obtained from equations (2) and (3) by replacing the operators with the functions. In terms of Schwinger-bosons the theory is invariant under $U(1)$ gauge transformations

$$\varphi'_{j\sigma}(\tau) = \mathrm{e}^{\mathrm{i}\alpha_j(\tau)}\varphi_{j\sigma}(\tau); \qquad \varphi'^\dagger_{j\sigma}(\tau) = \mathrm{e}^{-\mathrm{i}\alpha_j(\tau)}\varphi^\dagger_{j\sigma}(\tau) \tag{6}$$

with parameters which are period functions of Matsubara time $\alpha_j(0) = \alpha_j(\beta)$. The measure for the Schwinger-bosons includes Dirac-δ functions that enforce the constraint (3):

$$D\mu(\varphi^\dagger, \varphi) = \prod_{i,\tau,\sigma} \frac{D\varphi_{i\sigma}^\dagger(\tau) D\varphi_{i\sigma}(\tau)}{2\pi\mathrm{i}} \times \prod_{i\tau} \delta\left(\varphi_{i\sigma}^\dagger(\tau)\varphi_{i\sigma}(\tau) - 2S \right). \tag{7}$$

We introduce two spin-singlet Fermi fields

$$\Psi_i^{\mathrm{A}}(\tau) = \frac{1}{\sqrt{2S}} \varphi_{i\sigma}^\dagger(\tau) c_{i\sigma}(\tau), \tag{8}$$

$$\Psi_i^{\mathrm{B}}(\tau) = \frac{1}{\sqrt{2S}} \left[\varphi_{i1}(\tau)c_{i2}(\tau) - \varphi_{i2}(\tau)c_{i1}(\tau) \right], \tag{9}$$

which are gauge variant with charge -1 and 1 respectively:

$$\Psi'^{\mathrm{A}}_j(\tau) = \mathrm{e}^{-\mathrm{i}\alpha_j(\tau)}\Psi^{\mathrm{A}}_j(\tau), \qquad \Psi'^{\mathrm{B}}_j(\tau) = \mathrm{e}^{\mathrm{i}\alpha_j(\tau)}\Psi^{\mathrm{B}}_j(\tau). \tag{10}$$

The equations (8) and (9) can be regarded as an $SU(2)$ transformation:

$$\Psi_{i\sigma} = g^\dagger_{i\sigma\sigma'} c_{i\sigma'} \Rightarrow g_i^\dagger = \frac{1}{\sqrt{2S}} \begin{pmatrix} \varphi_{i1}^\dagger & \varphi_{i2}^\dagger \\ -\varphi_{i2} & \varphi_{i1} \end{pmatrix} \tag{11}$$

with $\Psi_i^{\mathrm{A}} = \Psi_{1i}$ and $\Psi_i^{\mathrm{B}} = \Psi_{2i}$. For that reason the Fermi measure is invariant under the change of variables. In terms of the spin-singlet Fermi fields the spin of the conduction electrons $\mathbf{s}_i$ has the form

$$s_i^\mu = \tfrac{1}{2} c_{i\sigma}^\dagger \tau^\mu_{\sigma\sigma'} c^\dagger_{i\sigma'} = \tfrac{1}{2} \mathrm{O}_i^{\mu\nu} \Psi^\dagger_{i\sigma} \tau^\nu_{\sigma\sigma'} \Psi_{i\sigma'}, \tag{12}$$

where

$$\mathrm{O}_i^{\mu\nu} = \tfrac{1}{2} \operatorname{Tr} g_i^\dagger \tau^\mu g_i \tau^\nu. \tag{13}$$

It is convenient to introduce three basic vectors which depend on the Schwinger-bosons:

$$T^1_{i\mu} = \mathrm{O}_i^{\mu 1} \qquad T^2_{i\mu} = \mathrm{O}_i^{\mu 2} \qquad T^3_{i\mu} = \mathrm{O}_i^{\mu 3}, \tag{14}$$

where $\mathbf{T}_i^3 = \frac{1}{S}\mathbf{S}_i$. Then, the spin of the electrons can be represented as a linear combination of three vectors $\mathbf{S}_j$, $\mathbf{P}_j = \mathbf{T}_j^1 + \mathrm{i}\mathbf{T}_j^2$ and $\mathbf{P}_j^\dagger = \mathbf{T}_j^1 - \mathrm{i}\mathbf{T}_j^2$:

$$\mathbf{s}_i = \frac{1}{2S}\mathbf{S}_i(\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{A}} - \Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{B}}) + \frac{1}{2}\mathbf{P}_i\Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{A}} + \frac{1}{2}\mathbf{P}_i^\dagger\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{B}}. \tag{15}$$

The basic vectors satisfy the relations $\mathbf{S}_i^2 = S^2$, $\mathbf{P}_i^2 = \mathbf{P}_i^{\dagger 2} = \mathbf{S}_i \cdot \mathbf{P}_i = \mathbf{S}_i \cdot \mathbf{P}_i^\dagger = 0$, and $\mathbf{P}_i^\dagger \cdot \mathbf{P}_i = 2$. Using the expression (15) for the spin of itinerant electrons, the total spin of the system $\mathbf{S}_i^{\mathrm{tot}} = \mathbf{S}_i + \mathbf{s}_i$ can be written in the form

$$\mathbf{S}_i^{\mathrm{tot}} = \frac{1}{S}\left[S + \frac{1}{2}\left(\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{A}} - \Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{B}} \right) \right] \mathbf{S}_i + \frac{1}{2}\mathbf{P}_i\Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{A}} + \frac{1}{2}\mathbf{P}_i^\dagger\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{B}}. \tag{16}$$

The gauge invariance imposes the conditions $\langle \Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{B}}\rangle = \langle \Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{A}}\rangle = 0$. As a result, the dimensionless magnetization per lattice site $M = \langle (S_i^{\mathrm{tot}})^z\rangle$ reads

$$M = \frac{1}{S}\left[S + \frac{1}{2}\left\langle\left(\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{A}} - \Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{B}}\right)\right\rangle\right]\langle \mathbf{S}_i^z\rangle. \tag{17}$$

At zero temperature $\langle \mathbf{S}_i^z\rangle = S$ and $M = S + m$, where

$$m = \tfrac{1}{2}\left\langle\left(\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{A}} - \Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{B}}\right)\right\rangle \tag{18}$$

is the contribution of the itinerant electrons.

Let us average the total spin of the system (equation (16)) in the subspace of the itinerant electrons $\langle \mathbf{S}_i^{\mathrm{tot}}\rangle_{\mathrm{f}} = \mathbf{M}_i$. In the path integral formalism this means integrating over the fermions. Then, the vector $\mathbf{M}_i$ ($\mathbf{M}_i^2 = M^2$) identifies the local orientation of the total magnetization. Accounting for the gauge invariance, one obtains an expression for $\mathbf{M}_i$ in terms of core spins $\mathbf{S}_i$:

$$\langle \mathbf{S}_i^{\mathrm{tot}}\rangle_{\mathrm{f}} = \mathbf{M}_i = \frac{M}{S}\mathbf{S}_i. \tag{19}$$

Now, if we use the Holstein–Primakoff representation for the vectors $\mathbf{M}_j$,

$$\begin{aligned} M_j^+ &= M_{j1} + \mathrm{i}M_{j2} = \sqrt{2M - a_j^\dagger a_j}\; a_j \\ M_j^- &= M_{j1} - \mathrm{i}M_{j2} = a_j^\dagger \sqrt{2M - a_j^\dagger a_j} \\ M_j^3 &= M - a_j^\dagger a_j \end{aligned} \tag{20}$$

the Bose fields a_j and $a_j^\dagger$ are the *true magnons* in the system. In terms of the true magnons the Schwinger-bosons (3) have the following representation:

$$\varphi_{i1} = \sqrt{2S - \frac{S}{M}a_i^\dagger a_i}, \qquad \varphi_{i2} = \sqrt{\frac{S}{M}}\, a_i. \tag{21}$$

Replacing the Schwinger-bosons in equations (8) and (9) for the spin-singlet fermions and keeping only the first two terms in $1/M$ expansion $\sqrt{1 - \frac{1}{2M}\, a_i^\dagger a_i} \simeq 1 - \frac{1}{4M}\, a_i^\dagger a_i + \cdots$, we obtain

$$\Psi_i^{\mathrm{A}} = c_{i1} + \frac{1}{\sqrt{2M}}\, a_i^\dagger c_{i2} - \frac{1}{4M} a_i^\dagger a_i c_{i1} + \cdots, \tag{22}$$

$$\Psi_i^{\mathrm{B}} = c_{i2} - \frac{1}{\sqrt{2M}}\, a_i c_{i1} - \frac{1}{4M} a_i^\dagger a_i c_{i2} + \cdots. \tag{23}$$

The equations (22) and (23) show that the singlet fermions are electrons dressed by a virtual cloud of repeatedly emitted and reabsorbed magnons.

An important advantage of working with A and B fermions is the fact that in terms of these spin-singlet fields the spin-fermion interaction is in a diagonal form, the spin variables (magnons) are removed, and one accounts for it exactly:

$$\sum_i \mathbf{S}_i \cdot \mathbf{s}_i = \frac{S}{2}\sum_i [\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{A}} - \Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{B}}]. \tag{24}$$

To proceed we rewrite the action (5) as a function of Schwinger-bosons and spin-singlet fermions:

$$\begin{aligned} A = \int_0^\beta \mathrm{d}\tau \Bigg[& \varphi_{i\sigma}^\dagger \dot{\varphi}_{i\sigma} + \Psi_i^{\mathrm{A}\dagger}\left(\frac{\partial}{\partial\tau} + \frac{1}{2S}\varphi_{i\sigma}^+\dot{\varphi}_{i\sigma}\right)\Psi_i^{\mathrm{A}} + \Psi_i^{\mathrm{B}\dagger}\left(\frac{\partial}{\partial\tau} - \frac{1}{2S}\varphi_{i\sigma}^\dagger\dot{\varphi}_{i\sigma}\right)\Psi_i^{\mathrm{B}} \\ & + \frac{1}{2S}\left(-\varphi_{i1}^\dagger\dot{\varphi}_{i2}^\dagger + \varphi_{i2}^\dagger\dot{\varphi}_{i1}^\dagger\right)\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{B}} + \frac{1}{2S}\left(-\varphi_{i2}\dot{\varphi}_{i1} + \varphi_{i1}\dot{\varphi}_{i2}\right)\Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{A}} \\ & + h\left(\varphi^\dagger, \varphi, \Psi^\dagger, \Psi\right)\Bigg]. \end{aligned} \tag{25}$$

It is convenient to write the Hamiltonian $h(\varphi^\dagger, \varphi, \Psi^\dagger, \Psi)$ as a sum of three terms

$$h = h_{\rm f} + h_{\rm H} + h_{\rm int}, \tag{26}$$

where $h_{\rm f}$ is the free **A** and **B** fermions' Hamiltonian,

$$h_{\rm f} = -t \sum_{\langle ij \rangle} \left(\Psi^\dagger_{i\sigma} \Psi_{j\sigma} + {\rm h.c.} \right) - \mu \sum_i \Psi^\dagger_{i\sigma} \Psi_{i\sigma} - \frac{J_{\rm H} S}{2} \sum_i \left(\Psi_i^{{\rm A}\dagger} \Psi_i^{\rm A} - \Psi_i^{+{\rm B}} \Psi_i^{\rm B} \right), \tag{27}$$

$h_{\rm H}$ is the Hamiltonian of the Heisenberg theory of antiferromagnetism (2), and $h_{\rm int}$ is the Hamiltonian of magnon–fermion interaction,

$$h_{\rm int} = -t \sum_{\langle ij \rangle} \left[\left[\frac{1}{2S} \left(\varphi^\dagger_{i\sigma} \varphi_{j\sigma} - 2S \right) \Psi^\dagger_{i\sigma'} \Psi_{j\sigma'} + {\rm h.c.} \right] \right.$$
$$\left. + \left[\frac{1}{2S} \left(\varphi^\dagger_{i1} \varphi^\dagger_{j2} - \varphi^\dagger_{j1} \varphi^\dagger_{i2} \right) \left(\Psi_j^{{\rm A}\dagger} \Psi_i^{\rm B} - \Psi_i^{{\rm A}\dagger} \Psi_j^{\rm B} \right) + {\rm h.c.} \right] \right]. \tag{28}$$

The action (25) is quadratic with respect to the spin-singlet fermions and one can integrate them out. We can accomplish this by first using the representation (21) of the Schwinger-bosons, then keeping only the quadratic terms with respect to the magnons, and finally calculating the diagrams in the leading order of gradient expansion. The action of the effective theory, in Gaussian approximation, is

$$A_{\rm eff} = \int_0^\beta {\rm d}\tau \left[a_i^\dagger \dot{a}_i + MJ \sum_{\langle ij \rangle} \left(a_i^\dagger a_i + a_j^\dagger a_j - a_i^\dagger a_j - a_j^\dagger a_i \right) \right], \tag{29}$$

where M is the dimensionless magnetization per lattice site (equation (17)) at zero temperature, and J is the effective exchange coupling:

$$J = -\frac{S^2}{M^2} J_{\rm AF} + \frac{t}{6M^2} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \frac{{\rm d}^3 k}{(2\pi)^3} \left(\sum_{\mu=1}^{3} \cos k_\mu \right) \left(n_k^{\rm A} + n_k^{\rm B} \right)$$
$$- \frac{2t^2}{3M^2 J_{\rm H} S} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \frac{{\rm d}^3 k}{(2\pi)^3} \left(\sum_{\mu=1}^{3} \sin^2 k_\mu \right) \left(n_k^{\rm A} - n_k^{\rm B} \right). \tag{30}$$

In equation (30), $n_k^{\rm R} = \theta(-\varepsilon_k^{\rm R})$ (R = A, B) are the occupation numbers for the A and B fermions with dispersions

$$\varepsilon_k^{\rm A} = -2t \left(\cos k_x + \cos k_y + \cos k_z \right) - \mu - \frac{J_{\rm H} S}{2}$$
$$\varepsilon_k^{\rm B} = -2t \left(\cos k_x + \cos k_y + \cos k_z \right) - \mu + \frac{J_{\rm H} S}{2}. \tag{31}$$

The first term in equation (30) comes from 'tadpole' diagrams with one A or B fermion line with vertices which relate to the first term in the Hamiltonian of interaction (28). The second term is obtained calculating the one-loop diagrams with A and B fermion lines, and with vertices which relate to the second term in $h_{\rm int}$. The term with time derivative in the effective action (29) is obtained by summing two terms. The first one is the term with time derivative in the action (25) which in terms of magnons has the form $\int_0^\beta {\rm d}\tau \frac{S}{M} a_i^\dagger \dot{a}_i$, while the second results from 'tadpole' diagrams with vertices related to the second and third terms of the action (25).

Based on the rotational symmetry, one supplements the action (29) up to an effective Heisenberg-like action, written in terms of the vectors $\mathbf{M}_i$:

$$H_{\rm eff} = -J \sum_{\langle ij \rangle} \mathbf{M}_i \cdot \mathbf{M}_j. \tag{32}$$

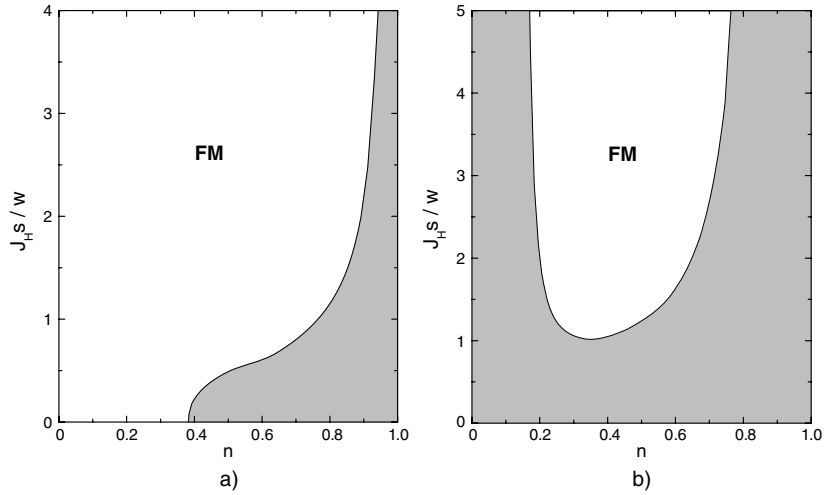

Figure 3. Phase diagrams when (a) $J_{\mathrm{AF}} = 0$ and (b) $\frac{J_{\mathrm{AF}}}{W} = 0.1$.

The ferromagnetic phase is stable if the effective exchange coupling constant is positive, $J > 0$. The dimensionless constant J/W, where $W = 12t$ is the band width, depends on J_{AF}/W, $J_{\mathrm{H}}S/W$ and $\mu/2t$. The $(\frac{J_{\mathrm{H}}S}{W}, n)$ phase diagram, where n is the carrier density, is depicted in figure 3 for $J_{\mathrm{AF}} = 0$ and $\frac{J_{\mathrm{AF}}}{W} = 0.1$.

The phase diagram shown in figure 3(a) ($J_{\mathrm{AF}} = 0$) is in a good agreement with phase diagrams obtained numerically [9] and by means of alternative analytical calculations [12]. Phase diagram figure 3(b) shows that direct antiferromagnetic exchange suppresses the ferromagnetism at small values of carrier concentrations, which is a well known experimental fact for manganites [8].

3. Canted ferromagnetism

After considering the pure double-exchange model, let us address the double-exchange model supplemented with on-site Coulomb repulsion (Hubbard term).

$$h = H - \mu N = -t \sum_{\langle ij \rangle} \left(c^{\dagger}_{i\sigma} c_{j\sigma} + \mathrm{h.c.} \right) - J_{\mathrm{H}} \sum_i \mathbf{S}_i \cdot \mathbf{s}_i + U \sum_i n_{i\uparrow} n_{i\downarrow} - \mu \sum_i n_i, \qquad (33)$$

where $n_{i\sigma} = c^{\dagger}_{i\sigma} c_{i\sigma}$. Our purpose is to show that canted ferromagnetism, with on-site spins of localized and delocalized electrons misaligned, emerges in the double-exchange model when the Coulomb repulsion is large enough.

Let us average the spin of the electrons (equation (15)) in the subspace of the itinerant electrons. We obtain, as a consequence of gauge invariance, that the spins of electrons are parallel to the localized spins $\langle \mathbf{s}_i \rangle_{\mathrm{f}} = \frac{m}{S} \mathbf{S}_i$. The order parameter of the rotational symmetry breaking is the vector $\langle \mathbf{S}^{\mathrm{tot}}_i \rangle$. In the ferromagnetic phase the spins of local and itinerant electrons are parallel, the ground state is infinitely degenerated, and one can choose the order parameter along the z-axis ($\langle \mathbf{S}^{\mathrm{tot}}_{3i} \rangle \neq 0$). When the on-site spins are misaligned the order parameter has two non-zero components: a component along the localized spin and a component perpendicular to it. Equation (15) shows that the on-site spins are misaligned if $\langle \Psi^{\mathrm{A}\dagger}_i \Psi^{\mathrm{B}}_i \rangle$ and $\langle \Psi^{\mathrm{B}\dagger}_i \Psi^{\mathrm{A}}_i \rangle$ are not equal to zero. To explore the on-site canted ferromagnetism we rewrite the Hamiltonian (33)

in terms of Schwinger-bosons and spin-singlet fermions. In particular, one obtains for the Hubbard term

$$\sum_i n_{i\uparrow} n_{i\downarrow} = -\sum_i \Psi_i^{\mathrm{A}\dagger} \Psi_i^{\mathrm{B}} \Psi_i^{\mathrm{B}\dagger} \Psi_i^{\mathrm{A}}. \tag{34}$$

We decouple this term by means of the Hubbard–Stratanovich transformation, introducing the complex field $\Delta_i(\Delta_i^\dagger)$, the projection of the order parameter on the plane perpendicular to the localized spin.

$$\mathrm{e}^{U\int_0^\beta \mathrm{d}\tau \sum_i \Psi_i^{\mathrm{A}\dagger}(\tau)\Psi_i^{\mathrm{B}}(\tau)\Psi_i^{\mathrm{B}\dagger}(\tau)\Psi_i^{\mathrm{A}}(\tau)} = \int \mathrm{d}\mu(\Delta^\dagger\Delta) \exp\left[-\int_0^\beta \mathrm{d}\tau \sum_i \left[\frac{\Delta_i^\dagger(\tau)\Delta_i(\tau)}{U} + \Psi_i^{\mathrm{A}\dagger}(\tau)\Psi_i^{\mathrm{B}}(\tau)\Delta_i(\tau) + \Delta_i^\dagger(\tau)\Psi_i^{\mathrm{B}\dagger}(\tau)\Psi_i^{\mathrm{A}}(\tau) \right] \right]. \tag{35}$$

Now, the partition function (4) can be represented as a path integral over the spin-singlet fermions, Schwinger-bosons, and complex order parameter. The integral over the fermions is Gaussian, and one can integrate them out. The resulting expression for the partition function is an integral over the Schwinger-bosons, and complex order parameter.

$$\mathcal{Z}(\beta) = \int \mathrm{d}\mu \left(\varphi^\dagger, \varphi, \Delta^\dagger, \Delta\right) \mathrm{e}^{-W\left(\varphi^\dagger, \varphi, \Delta^\dagger, \Delta\right)}. \tag{36}$$

We perform the integral over the collective variables $\Delta_i^\dagger$ and Δ_i using the steepest descent method. To this end, we set the spin fluctuation $a_i^\dagger$ and a_i (see equations (20), (21)) equal to zero and assume that the mean-field value of the order parameter $\Delta_i(\tau)$ is independent of τ and lattice sites i real constant Δ. Then, the free energy of the system, in mean-field approximation, is

$$\mathcal{F} = \frac{\Delta^2}{U} + \mathcal{F}_{\mathrm{f}}, \tag{37}$$

where $\mathcal{F}_{\mathrm{f}}$ is the free energy of a Fermi system with Hamiltonian

$$h_{\mathrm{f}} = \sum_k [\varepsilon_k^{\mathrm{A}} \Psi_k^{\mathrm{A}\dagger} \Psi_k^{\mathrm{A}} + \varepsilon_k^{\mathrm{B}} \Psi_k^{\mathrm{B}\dagger} \Psi_k^{\mathrm{B}} + \Delta(\Psi_k^{\mathrm{A}\dagger} \Psi_k^{\mathrm{B}} + \Psi_k^{\mathrm{B}\dagger} \Psi_k^{\mathrm{A}})]. \tag{38}$$

To write the Hamiltonian in diagonal form one introduces new Fermi fields ψ_k^a and ψ_k^b:

$$\Psi_k^{\mathrm{A}} = u\psi_k^a + v\psi_k^b, \qquad \Psi_k^{\mathrm{B}} = -v\psi_k^a + u\psi_k^b, \tag{39}$$

where the coefficients are

$$u = \sqrt{\frac{1}{2}\left(1 + \frac{J_{\mathrm{H}}S}{\sqrt{(J_{\mathrm{H}}S)^2 + 4\Delta^2}}\right)}, \qquad v = (\operatorname{sign}\Delta)\sqrt{1-u^2}. \tag{40}$$

Then,

$$h_{\mathrm{f}} = \sum_k \left[\varepsilon_k^a \psi_k^{a+} \psi_k^a + \varepsilon_k^b \psi_k^{b+} \psi_k^b\right]. \tag{41}$$

Here $\varepsilon_k^a = \varepsilon_k^-$, $\varepsilon_k^b = \varepsilon_k^+$, where

$$\varepsilon_k^\pm = \varepsilon_k \pm \tfrac{1}{2}\sqrt{(J_{\mathrm{H}}S)^2 + 4\Delta^2}, \tag{42}$$

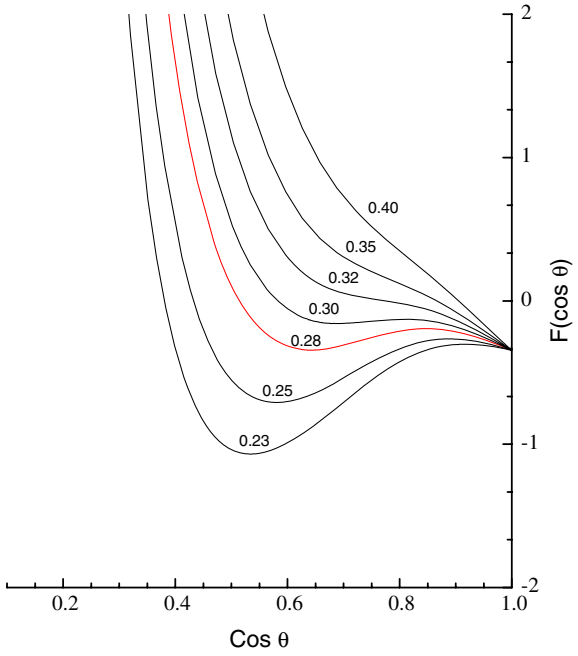

Figure 4. Dimensionless mean-field free energy $F = 6\mathcal{F}/W$, where W is the band width, as a function of $\cos\theta$, where θ is the angle between the on-site spin of carrier and localized spin, for $J_{\mathrm{H}}S/W = 2.32$, $\mu/W = -1.22$, and $W/U = 0.23$; 0.25; 0.28; 0.30; 0.32; 0.35; 0.40 (U: Coulomb repulsion).

and $\varepsilon_k = -2t[\cos(k_x)+\cos(k_y)+\cos(k_z)]-\mu$. Now, we can obtain the mean-field expression for the free energy. At zero temperature it is

$$\mathcal{F} = \frac{\Delta^2}{U} + \int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\frac{\mathrm{d}^3k}{(2\pi)^3}\left[\varepsilon_k^a\theta\left(-\varepsilon_k^a\right)+\varepsilon_k^b\theta\left(-\varepsilon_k^b\right)\right]. \tag{43}$$

It is convenient to introduce the angle between the on-site spin of the carrier and the localized spin:

$$\cos\Theta = \frac{\mathbf{S}_i\cdot\mathbf{s}_i}{|\mathbf{S}_i||\mathbf{s}_i|}. \tag{44}$$

Using equation (15) for the spin of itinerant electrons, and the properties of the basic vectors, one obtains

$$\cos\Theta = \left[1+\frac{4\langle\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{B}}\rangle\langle\Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{A}}\rangle}{\langle\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{A}}-\Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{B}}\rangle}\right]^{-\frac{1}{2}}. \tag{45}$$

We calculate the matrix elements in the formula (45) in mean-field approximation applying the transformation (39), (40). The result is

$$\begin{aligned}\langle\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{A}}-\Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{B}}\rangle &= \frac{J_{\mathrm{H}}S\left(n^a-n^b\right)}{\sqrt{(J_{\mathrm{H}}S)^2+4\Delta^2}}\\ \langle\Psi_i^{\mathrm{A}\dagger}\Psi_i^{\mathrm{B}}\rangle = \langle\Psi_i^{\mathrm{B}\dagger}\Psi_i^{\mathrm{A}}\rangle &= \frac{\Delta\left(n^a-n^b\right)}{\sqrt{(J_{\mathrm{H}}S)^2+4\Delta^2}},\end{aligned} \tag{46}$$

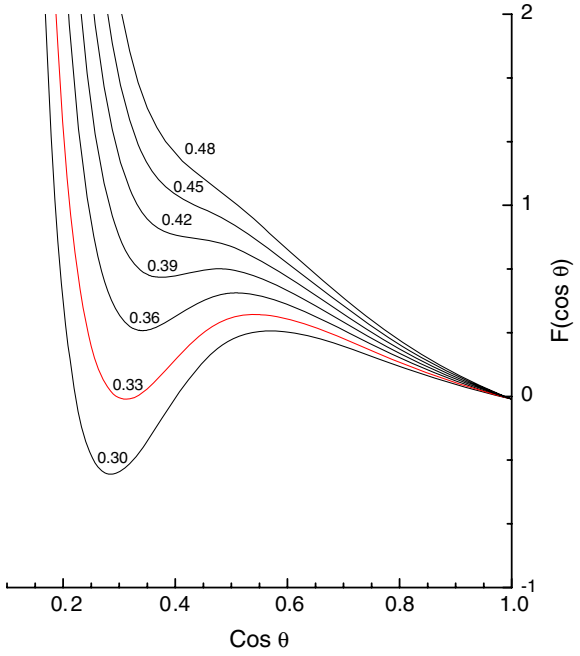

Figure 5. Dimensionless mean-field free energy $F = 6\mathcal{F}/W$, where W is the bandwidth, as a function of $\cos\theta$, where θ is the angle between the on-site spin of carrier and localized spin, for $J_{\mathrm{H}}\,S/W = 0.95$, $\mu/W = -5$, and $W/U = 0.30$; 0.33; 0.36; 0.39; 0.42; 0.45; 0.48 (U: Coulomb repulsion).

where n^a and n^b are occupation numbers for 'a' and 'b' fermions introduced by the transformation (39). After some algebra we arrive at the mean-field expression for the angle:

$$\cos\Theta = \frac{J_{\mathrm{H}}S}{\sqrt{(J_{\mathrm{H}}S)^2 + 4\Delta^2}}. \tag{47}$$

Next, we replace Δ in equations (42) and (43) by $\cos\Theta$ from equation (47) and rewrite the mean-field free energy as a function of $\cos\Theta$. The dimensionless energy $F = 6\mathcal{F}/W$ is depicted in figures 4–6 for different values of W/U and fixed $J_{\mathrm{H}}S/W$ and μ/W. As the graphs show, on increasing the Coulomb repulsion constant the system passes through a first-order quantum phase transition. Red lines (0.28—figure 4, 0.33—figure 5, and 0.77—figure 6) correspond to the critical values U_{c} of the Coulomb repulsion. The values U_{c} and Θ_{c} depend on the parameters of the theory such as Hund's coupling constant, chemical potential and band width. The character of the transition also depends on the parameters in the theory. We see (figure 6) that when $J_{\mathrm{H}}S/W = 0.50$ and $\mu/W = -0.33$ for small values of Coulomb repulsion, $W/U = 1.50$, 1.10, the minimum of the free energy is at $\cos\Theta = 1$. Near the quantum phase transition $W/U = 0.77$ the ground state is highly degenerated, while below this critical value, for large enough U, the on-site canted ferromagnetic state, with $\cos\Theta < 1$, is the ground state of the system. This could be an indication that near the quantum phase transition phase separation interpolates between ferromagnetic and on-site canted ferromagnetic order.

4. Conclusions

We have argued that the on-site Coulomb repulsion strongly affects the magnetic properties of spin-fermion systems. In particular, when the Coulomb repulsion is strong enough, the on-site

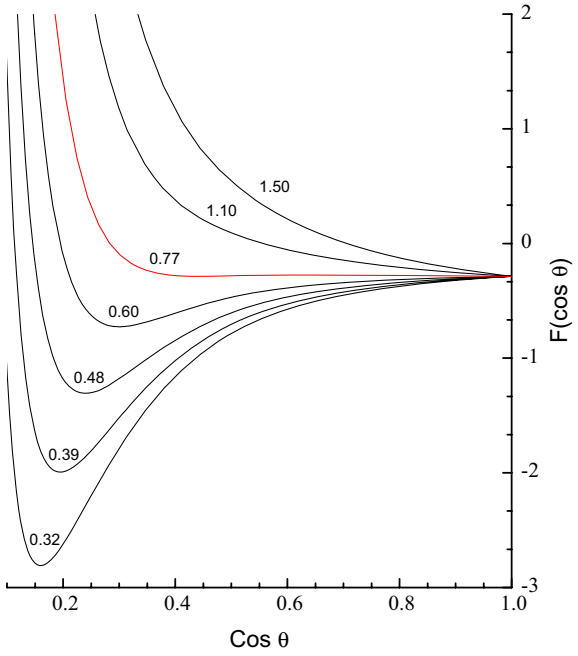

Figure 6. Dimensionless mean-field free energy $F = 6\mathcal{F}/W$, where W is the bandwidth, as a function of $\cos\theta$, where θ is the angle between the on-site spin of carrier and localized spin, for $J_{\mathrm{H}}\,S/W = 0.50$, $\mu/W = -0.33$, and $W/U = 0.32$; 0.39; 0.48; 0.60; 0.77; 1.10; 1.50 (U: Coulomb repulsion).

localized and carrier spins become misaligned (on-site canted ferromagnetic state). As follows from equation (47), $\cos\Theta > 0$ when $J_N > 0$ (see figure 2(a)), and $\cos\Theta < 0$ when $J_N < 0$ (see figure 2(b)). To obtain this result a double-exchange model with Hubbard term was considered. We represented the localized spins by means of Schwinger-bosons and introduced two spin-singlet fermion operators. In terms of the new Fermi fields the on-site Hund's interaction is in a diagonal form and the true magnons of the system can be recognized. Written in terms of Schwinger-bosons and spin-singlet fermions, the theory is $U(1)$ gauge invariant. We have shown that the on-site canted ferromagnetic state is a state with spontaneously broken gauge symmetry. This is because the order parameter is a gauge-varying collective field with charge -2 (see equations (10) and (35)):

$$\Delta'_j(\tau) = \mathrm{e}^{-\mathrm{i}2\alpha_j(\tau)}\Delta_j(\tau), \tag{48}$$

and a non-zero expectation value $\langle\Delta_j(\tau)\rangle \neq 0$ means spontaneous breakdown of the gauge symmetry.

To study the Goldstone modes in the on-site canted ferromagnetic phase it is convenient to represent the Schwinger-bosons and the order parameter in the form

$$\varphi_{j1} = \mathrm{e}^{\mathrm{i}\phi_j}\sqrt{2S - \frac{S}{M}a_j^\dagger a_j}, \qquad \varphi_{j2} = \sqrt{\frac{S}{M}}\,a_j, \qquad \Delta_j = |\Delta_j|\mathrm{e}^{\mathrm{i}\chi_j}. \tag{49}$$

Then the new fields, $a_j^\dagger, a_j, \phi_j$ and χ_j, transform under the gauge transformation in the following way:

$$a'_j = \mathrm{e}^{\mathrm{i}\alpha_j}a_j, \qquad \phi'_j = \phi_j + \alpha_j, \qquad \chi'_j = \chi_j - 2\alpha_j. \tag{50}$$

The theory is an $U(1)$ gauge theory and we have to impose one gauge-fixing condition. Hence, there are two Goldstone modes in the theory. For example, one can use $\phi_i = 0$ as a gauge-fixing condition (see equation (21)); then the Goldstone modes are the magnons a_i ($a_i^\dagger$) and χ_i phase. The physical origin of the extra mode is the totally broken rotation symmetry, while the mathematical reason is the spontaneous breakdown of the gauge symmetry. Alternatively, one can choose $\chi_i = 0$ for the gauge fixing. Both these conditions are not convenient. In the quadratic parts of the corresponding effective theories there are terms which mix magnons and phases. These terms are an obstacle to recognizing the spectrum in the theory. One hopes that there is a gauge-fixing condition which involves all gauge-varying fields and that the quadratic terms of the effective theory take a diagonal form. This issue will be addressed elsewhere.

In conclusion, it is important to stress that misalignment of the on-site localized and carrier spins is possible not only in the ferromagnetic phase. This is a characteristic feature of spin-fermion systems and one can consider the on-site canted antiferromagnetism or the on-site canted spiral phase if the theory parameters are appropriately chosen. The phase diagram which complements the known diagram of the DEM in the presence of strong Coulomb repulsion will be published elsewhere.

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