



Ising and spin orders in the iron-based superconductors

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Motivated by recent neutron scattering experiments, we study the ordering of spins in the iron-based superconductors $\text{LaFeAs}(\text{O}_{1-x}\text{F}_x)$, assuming them in proximity to a Mott insulator in the phase diagram. The ground state of the parent system with $x=0$ is a spin-density wave with ordering wave vector $\vec{Q}=(0, \pi)$ or $(\pi, 0)$. Upon raising the temperature, we find that the system restores $\text{SU}(2)$ symmetry, while an Ising symmetry remains broken, explaining the experimentally observed lattice distortion to a monoclinic crystal structure. Upon further temperature increase, the spins finally disorder at a second transition. The phase transition driven by doping with charge carriers similarly splits into an $\text{O}(3)$ transition and an Ising transition with $z=3$ at larger doping.

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After more than two decades of prevailing in condensed-matter physics, copper-based high-temperature superconductors have very recently given in to their iron-based cousins.¹⁻⁷ The newly discovered materials $M\text{FeAs}(\text{O}_{1-x}\text{F}_x)$, with M being rare earths such as La and Sm, have similar layered structure with stacked FeAs planes, sandwiched with La and O. Transport measurements show that the ground state of the undoped parent system is not an insulator, and local-density approximation (LDA) calculations have identified both a small electron pocket and a hole pocket at the Fermi level.⁸ However, it has been argued that the system is actually close to a Mott insulator, and a lot of physics can be studied in a similar manner as the copper-based high- T_c family, especially in the undoped system.⁹ In our current work, we will study the magnetism of these materials. Although the true unit cell of the FeAs plane contains two Fe ions, because of the staggered out-of-plane distribution of As ions (Fig. 1), we are only interested in the magnetic Fe ions and so we will use a unit cell with one Fe ion, unless stated otherwise.

Recent neutron scattering experiments have shown that by lowering temperature, the undoped material first undergoes a structural phase transition at 150 K, with a distortion from tetragonal structure to monoclinic structure, followed by a spin-ordering phase transition at 134 K developing stripe order at $(\pi, 0)$.¹⁰ The observed lattice distortion and spin-density wave (SDW) pattern are depicted in Fig. 1. In the superconducting material with $x=0.08$, the SDW order is not observed and, surprisingly, the distortion is absent as well, which suggests that the lattice distortion is driven by the development of the spin order. Interestingly, however, the SDW order at low doping only appears at a lower temperature (134 K) than the lattice distortion (150 K). We argue here that the SDW ordering is preceded, both in temperature and doping, by the breaking of an Ising symmetry in the effective spin model, and that this is responsible for the observed lattice distortion.

Reference 9 argued that the undoped material is described by either an $S=1$ or 2 spin model with nearest- and next-nearest-neighbor couplings J_1, J_2 that depend on the competition between the on-site Hubbard interaction and the Hund's rule,

$$H = \sum_{\langle i,j \rangle} J_1 \vec{S}_i \cdot \vec{S}_j + \sum_{\langle\langle i,j \rangle\rangle} J_2 \vec{S}_i \cdot \vec{S}_j. \quad (1)$$

Upon doping, this J_1 - J_2 model has $d_{x^2-y^2} + id_{xy}$ and then d_{xy} superconductivity as J_2 increases into the regime where the insulator has $(\pi, 0)$ SDW order.¹¹ There is also a much weaker interlayer coupling J_{\perp} , which is necessary to stabilize the spin order. It was suggested by first-principles calculations that both J_1 and J_2 are large and antiferromagnetic,¹² and Ref. 13 showed that $J_2 \sim 2J_1$. It is well known that when $J_1 < 2J_2$, the classical ground-state manifold of model (1) is $S^2 \otimes S^2$, because the two sublattices of the square lattice will each form a Néel order (\vec{n}_1 and \vec{n}_2), and the ground-state energy is independent of the relative angle between these two

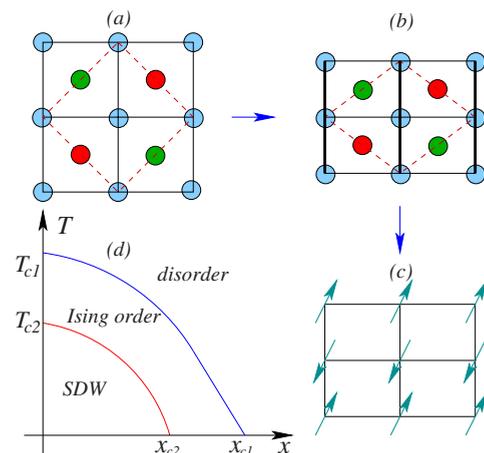


FIG. 1. (Color online) (a) The lattice structure at room temperature. The gray circles are Fe ions; the green (medium gray) and red (dark gray) circles are As ions above and below the Fe plane, respectively. The dashed square is the two-Fe unit cell. (b) The lattice structure between 150 and 134 K after the Ising order is developed: the thick lines represent the bonds between antiparallel aligned spins, but no uniform spin order is formed. The one-Fe unit cell is orthorhombic, but the two-Fe unit cell has a 3D monoclinic structure, as was seen in Ref. 10. (c) The $(0, \pi)$ spin order below 134 K. (d) The global phase diagram as a function of temperature and doping x . The blue (light gray) curve represents the Ising transition, the red (dark gray) curve represents the $\text{O}(3)$ transition.

Néel vectors. However, quantum or thermal fluctuations lift the degeneracy, leading to parallel or antiparallel alignment of the two sublattice Néel vectors.^{14–16} If we define O(3) vectors $\vec{\phi}_i$ as \vec{n}_i with softened unit-length constraint, the long wavelength field theory reads

$$L = \sum_{a=1}^2 \sum_{\mu=x,y} \partial_{\mu} \vec{\phi}_a \cdot \partial_{\mu} \vec{\phi}_a - r \vec{\phi}_a^2 + u (\vec{\phi}_a^2)^2 + L',$$

$$L' = \gamma \vec{\phi}_1 \partial_x \partial_y \cdot \vec{\phi}_2 - \alpha (\vec{\phi}_1 \cdot \vec{\phi}_2)^2. \quad (2)$$

In the above equation, we have absorbed the overall energy scale into $\vec{\phi}_a$. The parameter r is tuned by temperature, $\gamma \sim J_1/J_2$, α has contributions from both quantum and thermal fluctuations: $\alpha \sim J_1^2/J_2^2 \times (S\gamma_Q + \gamma_T T/J_2)$, and coefficients γ_Q and γ_T are given in Ref. 15. L' contains all sublattice couplings preserving the square lattice symmetry. The latter rules out the term $\vec{\phi}_1 \cdot \vec{\phi}_2$, but allows for the coupling $(\vec{\phi}_1 \cdot \vec{\phi}_2)^2$.

The ground-state manifold of the field theory (2) is $S^2 \otimes Z_2$, and the Z_2 order can be described by the Hubbard-Stratonovich field Φ , which couples to $\vec{\phi}_1 \cdot \vec{\phi}_2$: $L' = -\Phi(\vec{\phi}_1 \cdot \vec{\phi}_2) + \Phi^2/(4\alpha)$. The ordered state with $\Phi=1$ ($\Phi=-1$) corresponds to the $(\pi, 0)$ [$(0, \pi)$] SDW order. States with Ising Φ order, but only short-range SDW order, first appeared^{16,17} in the quantum theory of H for $S=1, 2$. If the coupling α is relevant, an Ising variable σ can be introduced directly as $\vec{\phi}_2 = \sigma \vec{\phi}_1$. Reference 15 showed that thermal fluctuations renormalize the anisotropy mixing γ to zero at long wavelength, so that at large scales the Lagrangian (2) can also be viewed as the low-energy field theory of the following Ising-O(3) model on the square lattice: $H = \sum_{\langle i,j \rangle} J(1 + \sigma_i \sigma_j) \vec{n}_i \cdot \vec{n}_j$. The O(3) vector \vec{n} denotes either of \vec{n}_1 or \vec{n}_2 , and the coarse-grained mode of σ is precisely the Ising field Φ introduced before. The easy-plane version of the Ising-O(3) model, dubbed the Ising-XY model, has been used widely as an effective model for the fully frustrated XY model on the square lattice and the triangular lattice.^{18–24}

Note that the Ising order $\Phi \neq 0$ does not imply O(3) order; however, because the system is invariant under exchanging $\vec{\phi}_1$ and $\vec{\phi}_2$, an O(3) order in $\vec{\phi}_{1,2}$ implies Ising order. Therefore, the transition temperature of the Ising order is necessarily no lower than that of the O(3) symmetry breaking. If we consider a purely two-dimensional system, at finite temperature there is only a two-dimensional (2D) Ising transition separating an Ising ordered phase and a disordered phase since a uniform O(3) order cannot exist at finite temperature in dimensions smaller than 3. The transition temperature can be estimated roughly as $T_{c1}/(J_2\alpha) \sim \xi^2/a^2$, where ξ is the correlation length of the 2D O(3) order at the transition and ξ^2/a^2 is a factor gained from integrating out the O(3) order parameters. A more precise estimate of the Ising transition temperature for the J_1 - J_2 model can be found in Ref. 15, with T_{c1} given by $T = 0.13 \frac{J_1^2 S}{J_2} \frac{\xi(T)^2}{a^2}$, in the large- S limit. The Ising order breaks the $\pi/2$ rotation symmetry of the square lattice; indeed, an order parameter $\Phi=1$ implies that the spins tend to be aligned parallel along

x but antiparallel along y . This Ising order favors a lattice contraction in the y direction, i.e., toward the orthorhombic structure in Fig. 1. The lattice distortion thus exists even in the absence of a uniform O(3) order, but it necessarily requires the Ising order. A similar mechanism was proposed for the lattice distortion in the cuprates.²⁵

The interlayer coupling J_{\perp} will drive the 2D Ising transition to a 3D Ising transition, but since it is much weaker than the intralayer couplings, it will not move the transition temperature significantly. However, the interlayer coupling stabilizes an O(3) ordered phase at finite temperature. Assuming the interlayer coupling is small, the transition temperature can be estimated as follows: The correlation length of the 2D O(3) nonlinear sigma model scales as $\xi/a \sim \exp(2\pi/g)$, with $g \sim \frac{T}{J_2 m^2}$, where m is the magnetic moment of the SDW order observed at zero temperature in units of the Bohr magneton, which is empirically found to be only $m \sim 0.36$; the interlayer coupling J_{\perp} grows under renormalization, and becomes unperturbative when $J_{\perp}/(J_2 m^2) \xi^2/a^2 \sim 1$, which will lead to the transition temperature $T_{c2} \sim 4\pi J_2 m^2 / \ln(m^2 J_2 / J_{\perp})$.²⁶ J_2 is evaluated to be ~ 1000 K in Ref. 13. Using the transition temperature from Ref. 10, the interlayer coupling J_{\perp} is estimated to be of order $10^{-4} J_2 m^2$, which can be neglected as compared with other interactions, unless we are very close to a critical point. The small value of the moment m is probably due to quantum fluctuations at zero temperature, since the system can be close to quantum phase transitions. Close to but above T_c , the correlation length of the system scales like in the 3D O(3) universality class, but once $\xi_z/a \sim [(T - T_{c2})/J_{\perp}]^{-\nu}$ shrinks to 1, the system crosses over to two-dimensional critical behavior. The fact that the lattice distortion observed in experiments¹⁰ occurs at a temperature that is relatively small compared to the exchange interaction J_2 (Ref. 13) is probably due to the proximity to quantum phase transitions, which is consistent with the small magnetic moment observed at low temperature.¹⁰ The phase diagram is depicted in Fig. 1.

Quantum phase transitions. In $\text{LaFeAsO}_{1-x}\text{F}_x$, the SDW order vanishes as a small amount of extra carriers are introduced by doping, meanwhile the superconductor state emerges, implying the presence of one or more quantum phase transitions as a function of doping. A tentative quantum critical point in these systems has already been studied experimentally in a series of samples $\text{SmFeAsO}_{1-x}\text{F}_x$.²⁷ Since the nature of the superconductor is not yet clear, however, in the present work we focus on the quantum phase transitions of the spin system discarding the presence of superconductivity. In terms of the itinerant fermions, the SDW at $(\pi, 0)$ can be understood from the large susceptibility arising from the location of electron and hole pockets in the Brillouin zone: there are low-energy electron-hole pair excitations at the $(\pi, 0)$ wave vector (Fig. 2). As extra electrons are doped into this system, these low-energy excitations disappear rapidly because of the unequal sizes of the electron and hole pockets. The SDW order parameter cannot decay into a particle-hole pair excitations preserving both momentum and energy, because the SDW wave vector $(\pi, 0)$ does not connect two pairs of points on the Fermi surface for finite x (Fig. 2). After integrating out electrons, we would obtain the following $z=1$ Lagrangian:

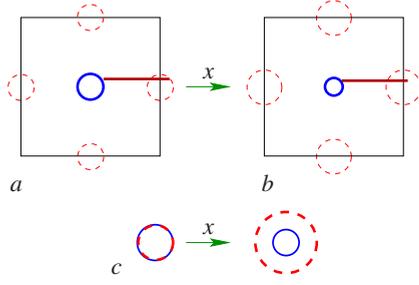


FIG. 2. (Color online) (a) The Brillouin zone for a one-Fe unit cell at zero doping. The thick blue (dark gray) circle denotes two almost overlapping hole pockets (Ref. 30), the red (light gray) dashed circle represents the electron pocket. The line that connects the electron and hole pockets is the $(\pi, 0)$ wave vector. (b) At finite doping, the electron pockets expand and the hole pockets shrink, so that the $(\pi, 0)$ vector can no longer connect points at the Fermi level. (c) Translation of the electron pocket by vector $(\pi, 0)$: If at zero doping there is a perfect overlap of the pockets, at infinitesimal doping there is no crossing between electron and hole Fermi level at all, i.e., an order with $(\pi, 0)$ wave vector cannot decay into a particle-hole pair.

$$L = \sum_{i=1}^2 \sum_{\mu=\tau,x,y} \partial_{\mu} \vec{\phi}_i \cdot \partial_{\mu} \vec{\phi}_i - r \vec{\phi}_i^2 + u |\vec{\phi}_i|^4 + L',$$

$$L' = \gamma \vec{\phi}_1 \cdot \partial_x \partial_y \cdot \vec{\phi}_2 + \gamma_1 |\vec{\phi}_1|^2 |\vec{\phi}_2|^2 - \alpha (\vec{\phi}_1 \cdot \vec{\phi}_2)^2, \quad (3)$$

which contains no dissipative term. The first three terms of the Lagrangian describe the two copies of 3D O(3) systems on the two sublattices. The first term in L' mixes $\vec{\phi}_1$ and $\vec{\phi}_2$, and its scaling dimension is

$$\Delta[\gamma] = D - (2 + D - 2 + \eta) = -\eta. \quad (4)$$

$\eta=0.0375$ (Ref. 28) is the anomalous dimension of $\vec{\phi}$ at the 3D O(3) universality class, therefore the γ term is irrelevant. The second term in L' is allowed by symmetry and hence will be generated under renormalization. Its scaling dimension can be evaluated as

$$\Delta[\gamma_1] = D - 2\Delta[|\vec{\phi}|^2] = D - 2\left(D - \frac{1}{\nu}\right) = \frac{2}{\nu} - D, \quad (5)$$

with the correlation length exponent $\nu=0.71$ for the 3D O(3) transition. The γ_1 term is thus also irrelevant. However, the α term is relevant at the 3D O(3) transition, since it has positive scaling dimension $\Delta[\alpha]=0.581$.²⁸ We expect this term to split the two coinciding O(3) transitions into two transitions, an O(3) transition and an Ising transition, as was found in the Schwinger boson theory.^{16,29} Again, because the O(3) order of $\vec{\phi}_{1,2}$ implies Ising order, the latter should occur after the O(3) transition, i.e., at larger x . The distance in doping between the two transitions can be estimated by scaling, ignoring possible higher-order singular perturbations mediated by electrons,

$$\Delta x \sim \frac{\Delta r_c}{r_c} \sim \alpha^{1/\nu\Delta[\alpha]} = \left(\frac{J_1}{J_2}\right)^{2/\nu\Delta[\alpha]}. \quad (6)$$

Note that the monopoles of $\vec{\phi}_1$ and $\vec{\phi}_2$ are confined by the α term. The Berry phase for monopoles of a spin- S system on the square lattice is proportional to $i\pi S$; the monopole-composite of ϕ_1 and ϕ_2 carries a trivial Berry phase for the $S=1$ and 2 cases,^{16,29} and hence is ignored hereafter.

Note that while the O(3) SDW order parameters $\vec{\phi}_1$ and $\vec{\phi}_2$ cannot decay into particle-hole excitations since the wave vector $(\pi, 0)$ does not connect pairs of points at the Fermi level, the same is not true for the Ising order parameter $\Phi \sim \vec{\phi}_1 \cdot \vec{\phi}_2$ which orders at $(0, 0)$. Also since Φ changes sign under a $\pi/2$ rotation and reflection along the axis $x=y$, but does not break any other symmetry, Φ couples to the two-body d -wave density $\Phi_q \sim \sum_k \text{sign}(k_x^2 - k_y^2) c_{k+q/2}^\dagger c_{k-q/2}$, and hence can decay into particle-hole excitations. The decay rate can be calculated using Fermi's golden rule,

$$\text{Im}[\chi(\omega, q)] \sim \int \frac{d^2k}{(2\pi)^2} [f(\epsilon_{k+q}) - f(\epsilon_k)] \delta(\omega - \epsilon_{k+q} + \epsilon_k) \times |k|\Phi_q|k+q|^2 \sim c_0 \frac{\omega}{q}. \quad (7)$$

The standard Hertz-Millis³¹ formalism leads to a $z=3$ theory with Lagrangian

$$L = \Phi_{-q} \left(\frac{|\omega|}{c_0 q} + c_1 q^2 + r \right) \Phi_q + \dots. \quad (8)$$

The ellipses stand for all the quartic and higher-order terms of Φ , which are irrelevant at this Gaussian fixed point described by Eq. (8). Quadratic terms with singular factor ω^2/q^2 or higher may occur in the expansion, but since the theory has $z=3$, these terms are irrelevant. The $z=3$ critical field theory was also obtained for the electronic nematic phase with an order parameter similar to Φ .³² The critical exponents can be extracted directly from the field theory (8). For instance, in the quantum critical region, the specific heat and the critical temperature of the finite temperature Ising transition scale as

$$C_v \sim T^{d/z} = T^{2/3},$$

$$T_{c1} \sim (x_{c1} - x)^{z/(d-2+z)} = x_{c1} - x, \quad (9)$$

with $d=2$. The weak interlayer coupling $w\Phi_n\Phi_{n+1}$ will finally drive the scaling back to three-dimensional behavior with $w \sim J_{\perp}/J_2$, but its role is not considerable unless the 2D correlation length is long enough, i.e., if we are close enough to the quantum critical point. The spatial scaling dimension of w is $\Delta[w]=2$ at the 2D critical point described by Eq. (8), therefore w becomes nonperturbative when $(\xi/a)^{\Delta[w]} \sim 1/w$, i.e.,

$$x_{c1} - x \sim w^{1/(\nu\Delta[w])} = w. \quad (10)$$

Within this small window, the critical scaling becomes

$$C_v \sim T^{d/z} = T,$$

$$T_c \sim (x_{c1} - x)^{z/(d-2+z)} = (x_{c1} - x)^{3/4}. \quad (11)$$

The O(3) order parameter $\vec{\phi}$, which can be taken as $\vec{\phi}_1$, cannot decay into particle-hole pairs, assuming the $(\pi, 0)$ wave vector does not connect two points at the Fermi level. The Gaussian part of the Lagrangian describing the O(3) transition at x_{c2} has dynamical exponent $z=1$,

$$L = \vec{\phi}_{-q}(\omega^2 + q^2)\vec{\phi}_q + L'. \quad (12)$$

L' consists of quartic and higher-order terms. If the quartic terms have no singularity in momentum and frequency space, the Lagrangian (12) describes a 3D O(3) transition. Berry phases of monopoles in this case are trivial for spin-1,2 (Refs. 16 and 29) and so are not noted. However, the quartic terms of the effective action may include singular terms like

$$L' = \gamma_2 |\vec{\phi}_{-q}^2| \frac{|\omega|}{q} |\vec{\phi}_q^2|. \quad (13)$$

This term can be viewed as describing the decay of $|\vec{\phi}|^2$, which couples to the zero momentum charge density. From naive power counting, γ_2 has the same scaling dimension as all the other quartic terms without singularities. However, since it mixes the $|\vec{\phi}|^2$ field at distinct spatial points, the anomalous dimensions will be contributed by the two different points separately. Therefore, its scaling dimension can be evaluated as $\Delta[\gamma_2] = D - 2(D - \frac{1}{\nu}) = \frac{2}{\nu} - D$, which is again irrelevant at the 3D O(3) transition. If no other more relevant

quartic terms are present, the quantum phase transition of $\vec{\phi}$ at x_{c2} belongs to the 3D O(3) universality class, cf. Fig. 1. But a thorough analysis of the quartic terms is required to draw a firm conclusion.

Quantum critical points play an important role in transport because the electrons can scatter off the critical modes. We expect the Ising critical modes to contribute the dominant part to the low-temperature resistivity, because of its $z=3$ soft modes and the ensuing larger density of states at low energy. At low temperature where the scattering is dominated by small-angle forward scattering, the resistivity is expected to scale as $\rho \sim T^{4/3}$. The more general formula for the resistivity for a $z=3$ theory with Lagrangian (8) reads $\rho \sim T^{(d+2)/z}$, which is consistent with the well-known $T^{5/3}$ law of the resistivity at the quantum critical point of three-dimensional itinerant ferromagnetic order.³³

In summary, we have studied the SDW at $(\pi, 0)$ observed experimentally in $\text{LaFeAsO}_{1-x}\text{F}_x$, and its phase transitions. While raising the thermal and quantum fluctuations, the SDW is predicted to cede to a state with restored SU(2) invariance, but retaining a broken Ising symmetry which drives a lattice distortion. This is followed by an Ising transition at higher temperature or larger doping. The nature and universality classes of these transitions and various critical exponents are discussed.

Note added. Fang *et al.*³⁴ have also applied thermal fluctuations of the J_1 - J_2 model to the iron-based superconductors.

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