Correction

PHYSICS

The authors note that in the acknowledgments section on page 4121, National Science Foundation Grant DMR-0706625 and Robert A. Welch Foundation Grant C-1411 were inadvertently omitted. The corrected acknowledgments section appears below.

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Iron pnictides as a new setting for quantum criticality

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Two major themes in the physics of condensed matter are quantum critical phenomena and unconventional superconductivity. These usually occur in the context of competing interactions in systems of strongly correlated electrons. All this interesting physics comes together in the behavior of the recently discovered iron pnictide compounds that have generated enormous interest because of their moderately high-temperature superconductivity. The ubiquity of antiferromagnetic ordering in their phase diagrams naturally raises the question of the relevance of magnetic quantum criticality, but the answer remains uncertain both theoretically and experimentally. Here, we show that the undoped iron pnictides feature a unique type of magnetic quantum critical point, which results from a competition between electronic localization and itinerancy. Our theory provides a mechanism to understand the experimentally observed variation of the ordered moment among the undoped iron pnictides. We suggest P substitution for As in the undoped iron pnictides as a means to access this example of magnetic quantum criticality in an unmasked fashion. Our findings point to the iron pnictides as a much-needed setting for quantum criticality, one that offers a unique set of control parameters.

Quantum Criticality in the Pnictides

The existence of the antiferromagnetic state naturally raises the possibility of carrier-doping-induced quantum phase transitions in the iron pnictides (9–11), but the situation is not yet certain. Theoretically, the evolution of the Fermi surface as a function of carrier doping is not yet well understood, and this limits the study of quantum criticality. Experimentally, earlier measurements in LaO1−xFeAs (1) and SmO1−xFeAs (12) show a moderate suppression of the magnetic/structural transition temperature(s) as x is increased; beyond x of about ~7%, the transitions are interrupted by superconductivity. Further experiments have led to conflicting reports for the first-order or second-order nature of the carrier-induced zero-temperature magnetic and structural phase transitions (13–15).

We propose that an alternative to a possible doping-induced quantum phase transition is one that is accessed by changing the relative strength of electron–electron correlations. Thus, we suggest that the iron pnictides may exhibit an example and setting for quantum criticality. Our approach is motivated by the phenomenological and theoretical evidence that the parent iron pnictide is a “bad metal” (9, 16, 17). Accordingly, we formulate our considerations in terms of an incipient Mott insulator: the electron-electron interactions lie close to, but do not exceed the critical value for the insulating state. Within this picture, the electronic excitations comprise an incoherent part away from the Fermi energy, and a coherent part in its vicinity. The incoherent electronic excitations are described in terms of localized Fe magnetic moments, with frustrating superexchange interactions. The latter have been discussed earlier by two of us (9) and others (18). This division of the electron spectrum is a simple and convenient way of analyzing the complex behavior of a bad metal close to the Mott transition, whose spectrum exhibits incipient upper and lower Hubbard bands and a coherent quasi-particle peak at the Fermi energy (19).

The coupling of the local moments to the coherent electronic excitations competes against the magnetic ordering. A magnetic quantum critical point arises when the spectral weight of the coherent electronic excitations is increased to some threshold value.

The Electron Spectrum

The incoherent and coherent parts of the single-electron spectral function are illustrated schematically in Fig. 1. The central peak describes the coherent itinerant carriers; these are the electronic excitations that are responsible for a Drude optical response and that are adiabatically connected to their noninteracting counterparts. The side peaks describe the incoherent excitations, vestiges of the lower and upper Hubbard bands associated with a Mott insulator that would arise if the electron–electron interactions were larger than the Mott localization threshold. Each of the three peaks may in general have a complex structure due to the multiorbital nature of the iron pnictides. The decomposition of the electronic spectral weight into coherent and incoherent parts is natural for a metal near a Mott transition (19, 20).

We use w to denote the percentage of the spectral weight lying in the coherent part of the spectrum. A relatively small w may be inferred for the iron pnictides, because the Drude weight seen in the optical conductivity (21–23) is very small (on the order of 5% of the total spectral weight integrated to ~2 eV). A small w corresponds to an interaction strength sufficiently large that the system is close to the Mott transition, albeit on the metallic side: this implies a large electron–electron scattering rate, consistent with the observed large electrical resistivity (on the order of 0.5 mΩ · cm for single crystals and 5 mΩ · cm for polycrystals) at room temperature. In terms of electrical conduction, the iron pnictides are similar to, e.g., V2O3, a bad metal (with a room temperature resistivity (24) of about 0.5 mΩ · cm) that is known to be on the verge of a Mott transition, and is very different from, e.g., Cr, a simple metal (with a room temperature resistivity (25) of ~0.01 mΩ · cm) which orders into a spin-density-wave ground state.

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Here, \( r \) labels the three Pauli matrices. In the projection procedure leading to Eq. 2, we keep \( d_{\text{coh}} \) as part of the low-energy degrees of freedom; the prefactor \( w \) in the first equation comes from the rescaling \( c_{\text{coh}} = (1/\sqrt{w}) d_{\text{coh}} \) and \( E_{\text{coh}} \) is therefore the conduction–electron dispersion at \( w = 1 \). At the same time, we integrate out the high-energy states involved in \( V_{\text{coh}} \). To the leading order in \( w \), this procedure is carried out at the \( w = 0 \) point which is taken to have a full gap (26); as a result, the effective coupling \( G_{\text{coh}} \) is of order \( w^{\beta} \). Beyond the leading order in \( w \), the coupling constants will acquire further corrections. The computation of these corrections is difficult, since, at those orders, the spectrum becomes continuous from the coherent to incoherent part (Fig. 1); it is left for future work. Still, our leading-order analysis captures the form of the low-energy effective Hamiltonian, which is

\[
H_{\text{eff}} = H_J + H_c + H_m.
\]

**J₁–J₂ Competition**

The superexchange interactions in the iron pnictides contain n.n. and n.n.n. terms because of the specific relative locations of the ligand As atoms and Fe atoms (9, 18, 27). To assess the tunability of \( J_1 \) and \( J_2 \), we consider an oversimplified case, illustrated in Fig. 2. Here, only one Fe 3d orbital is considered. We assume that the 3d orbital on each of the 4 corners of a square plaquette has an identical hybridization matrix element, \( V \), with one As 4p orbital located above the center of the plaquette. The superexchange interaction is found to be \( J_1 \propto \sum \langle \sum \delta_\text{2D} (r) \rangle \), where \( r \) labels a plaquette in the 2D square lattice and the summation \( \sum \) is over the 4 Fe sites of a plaquette. For classical spins, this is the canonical case of magnetic frustration: all states with \( \sum \delta_\text{2D} (r) = 0 \) are degenerate. Written in the form of Eq. 1, this corresponds to \( J_2 = J_1/2 \). This discussion is instructive for the understanding of the realistic exchange interactions in the iron pnictides. Several aspects are neglected in the simplified analysis given above. First, multiple 3d orbitals are important, and the hybridization is orbital-sensitive. Both the \( J_1 \) and \( J_2 \) interactions are therefore matrices. Second, the real band structures must be described by more complex \( p-p-p \) and \( d-d \) tight-binding parameters. Both features spoil the elementary \( J_2 = J_1/2 \) relationship. Still, the simple considerations given above suggest that the overall strength of \( J_2 \) and \( J_1 \), i.e., the largest eigenvalues of the \( J_1 \) and \( J_2 \) matrices, are comparable with each other. Detailed analysis of the matrix elements indicates that there are more entries in the \( J_2 \) matrix than in the \( J_1 \) matrix that correspond to the dominating antiferromagnetic component, and that the overall magnitude (the largest eigenvalue) of the \( J_2 \) matrix will be somewhat larger than half of that of the \( J_1 \) matrix. This conclusion is supported by the fitting of the ab initio results.
of the ground-state energies for various magnetic configurations in terms of $J_3$ and $J_4$ parameters in the (nonmatrix) Heisenberg form (18, 27). This range of $J_2/J_1$ leads to a two-sublattice collinear antiferromagnetic ground state, consistent with the results of the neutron scattering experiment (2).

On the one hand, the above argument implies that the magnetic frustration effect is strong, and can provide significant quantum fluctuations leading to a reduced ordered moment. On the other hand, it suggests that the degree to which $J_2/J_1$ can be tuned in practice could be limited.

**Magnetic Quantum Critical Point**

The order parameter for the two-sublattice antiferromagnet appropriate for $J_2/J_1 > 1/2$ is the staggered magnetization, $\mathbf{m}$, at wave vector $\mathbf{Q} = (\pi, 0)$. The effective theory for the $H_2$ term alone corresponds to a $\phi^4$ theory whose action is of the form $S \sim r \phi^2 + \mu \phi^4$. The coupling to the coherent quasi-particles is given by the $H_0$ term of Eq. 2; it causes a shift of the tuning parameter $r$ and also introduces a damping term. These contributions to the $\gamma$ coefficient are given by:

$$\Delta \gamma + \Gamma = \sum_{k,\alpha,\beta,\gamma} \frac{g_{\alpha\beta\gamma}}{\lambda_{\alpha\beta\gamma}} \frac{f(\mathbf{k}+\mathbf{q},\omega) - f(\mathbf{k},\omega)}{\lambda_{\alpha\beta\gamma} - (\mathbf{k}+\mathbf{q},\omega) - (\mathbf{k},\omega)} .$$

[Eq. 4]

Here, $f(\omega)$ is the Fermi–Dirac distribution function and $a_{\gamma}$ is an orbital-dependent coefficient: $\sum_{\gamma} a_{\gamma}$ appears in the order parameter for the $(\pi, 0)$ antiferromagnet. Note that both $g_{\alpha\beta\gamma}$ and $\epsilon_{\alpha\beta\gamma}$ are linear in $w$. We can infer from Eq. 4 that the damping term is of the order $w^2$ at low energies: for $|\omega| \ll W$ ($W$ is the bandwidth), $\gamma = \gamma_0 (\omega_0, \gamma_0)$, where $\gamma_0$ is, to leading order in $w$, the constant value associated with the couplings and density of states of the $w = 0$ case. Note that $\gamma$ is nonzero because, for the parent compounds, $Q$ connects the hole pockets near the $\Gamma$ point of the Brillouin zone (BZ) and the electron pockets near the M points (in the unfolded BZ notation). At the same time, $\gamma$ does not diverge since the nesting is not perfect. The existence of the linear in $\omega$ damping term is in contrast to the doped case, where $Q$ no longer connects the hole- and electron-Fermi surfaces (11). Importantly, we can also infer from Eq. 4 that the leading frequency- and temperature-independent term $\Delta \gamma + \Gamma = w_0 Q$ is linear in $w$, with $A_Q = \sum_{\alpha,\beta,\gamma} g_{\alpha\beta\gamma}^2 \lambda_{\alpha\beta\gamma}^2 \Theta(E_F - E_k + Q) - \Theta(E_F - E_k)/E_k (\Delta_\alpha\beta\gamma) \Theta(E_k - \mu_c, \mu_c + Q)$ (where $\Theta$ is the Heaviside function) is independent of $w$, and positive.

The low-energy Ginzburg–Landau theory then takes the form,

$$S = \int d\mathbf{q} \int d:\omega \frac{r(\mathbf{w}) + (\mathbf{q} - \mathbf{Q})^2}{w_0 Q} + \omega^2 + \gamma(\omega) \left[ \mathbf{m}(\mathbf{q}, \omega) \right]^2$$

$$+ w \int d\mathbf{q} \int d\omega \delta \left( \sum_i \mathbf{q}_i \right) \delta \left( \sum_i \omega_i \right) \left[ \mathbf{m} \right]^4 + \ldots$$

[Eq. 5] where $r(\mathbf{w}) = r(\mathbf{w} = 0) + w_0 Q$, $r(\mathbf{w} = 0)$ is negative, placing the linear in $w$ shift, $w_0 Q$, causes $r(\mathbf{w})$ to vanish at $\mathbf{w} = w_c$, leading to a quantum critical point. In terms of the external control parameter $\delta$, shown in Fig. 3, $w = w_c$ defines $\delta = \delta_c$. The $\phi^4$ theory describes $a = 2$ (where $z$ is the dynamical exponent) antiferromagnetic quantum phase transition, which is generically second order.

The $O(3)$ vector $\mathbf{m}$, corresponding to the $(\pi, 0)$ order, is accompanied by another $O(3)$ vector $\mathbf{m}'$ that describes the $(0, \pi)$ order. These two vector order parameters accommodate a composite scalar, $\mathbf{m} \cdot \mathbf{m}'$, the order parameter for an Ising transition (10, 11, 28). In turn, the Ginzburg–Landau action, Eq. 5, contains a quartic coupling $\tilde{u}(\mathbf{m} \cdot \mathbf{m})^2$ as well as $\tilde{w}(\mathbf{m}' \cdot \mathbf{m}'')^2$. In the $\mathbf{z} = 2$ case here, the $\phi^4$ theory is at effective dimension $d + \mathbf{z} = 4$. At the QCP of the $O(3)$ transition, the $\tilde{u}$ quartic coupling term is marginally relevant in the renormalization group sense. The $T = 0$ transition could therefore either be turned to first order, or be split into two continuous transitions, one for the Ising transition, whose scalar order parameter is $\tilde{m} = m$ (which corresponds to the structural distortion when it is coupled to some structural degrees of freedom), the other is for the $O(3)$ magnetic one. Either effect is expected to be weak, because of the marginal nature of the coupling.

The magnetic quantum criticality will strongly contribute to the electronic and magnetic properties in the quantum critical regime. We note that since $\mathbf{z} = 2$, there are (marginal) logarithmic corrections to simple Gaussian critical behavior (29). Following discussions in, e.g., ref. 29, we expect that the specific-heat coefficient will be $C / T \sim \ln(1/T)$, the NMR relaxation rate $1 / T_1 \propto c$, and (in the presence of disorder scattering that smears the Fermi surface) the resistivity $\rho \propto T$.\[fig 3\]

**Tuning Parameter and Variation of Magnetic Order**

The parent materials of the different iron arsenides will have different internal pressures and “$c/a$” ratios, and will correspondingly have different ratios of the electron–electron interaction to the effective bandwidth. According to our theory, the resulting variation of the coherent spectral weight $w$ will, in turn, tune the control parameter $r$ in Eq. 5, and the ordered moment will change accordingly across the different compounds.

Neutron scattering experiments have indeed found that the ordered moment does vary across the parent arsenides. The moment associated with Fe-ordering at low temperatures is $\approx 0.2-0.3 \mu_B / \text{Fe}$ in NdOFeAs (30, 31), $0.4 \mu_B / \text{Fe}$ in LaOFeAs (2), $0.5 \mu_B / \text{Fe}$ in PrOFeAs (32, 33), and $0.8-1.0 \mu_B / \text{Fe}$ in CeOFeAs (15), BaFe$_2$As$_2$ (34), and SrFe$_2$As$_2$ (35).

**$As_{1-x}P_x$ Series of the Parent Iron Phosphides**

Because the $c$-lattice constant in LaOFeP is smaller than that in LaOFeAs, these considerations suggest that the coherent-electron spectral weight of the iron phosphides is larger than that of the iron arsenides. A consequence is that, in contrast to the arsenide, the phosphide does not have a magnetic transition (36). We then propose that a parent iron pnictide series created by P doping of As presents a means to unmask a magnetic quantum critical point. Our purpose is better served the weaker the superconductivity is in the P end material. LaOFeAs$_{1-x}$P$_x$ is promising, since LaOFeP is a weak superconductor whose $T_c$ is only a few Kelvin or may
even vanish (37–39). CeOFeAs1−xP may also be of interest in this context. While CeOFeAs (15) is antiferromagnetic, CeFeP is a paramagnetic metal (40). We remark in passing that P-doping for As is more advantageous than external pressure, because the latter is known to cause a volume collapse (41). It would be interesting to search for a substitution for As such that w could be reduced, leading toward to the Mott insulating state.

To understand the tuning of the microscopic electronic parameters, we have carried out density-functional-theory (DFT) calculations on both CeOFeAs and CeFeP for comparison. We find that the d-p hybridization matrix is larger in CeOFeP than in CeOFeAs. This is consistent with the qualitative consideration that, compared with CeOFeAs, CeFeP has a higher internal pressure and, hence, a higher kinetic energy and smaller ratio of the interaction to the bandwidth, thus a larger coherent weight w.

**Comparison with DFT Studies**

We have considered the mechanism for quantum fluctuations having in mind the proximity to the Mott limit, where the instantaneous atomic moment. Quantum fluctuations will then lead to a reduced ordered moment in the true ground state. The J1−J2 competition together with the coupling of the local moments to the coherent itinerant electronic excitations arising naturally in the Mott-proximity picture we have described is just such a mechanism for quantum fluctuations.

**Discussion**

We have developed a framework to describe the magnetism of the iron pnictides, appropriate for electron–electron interactions that are of an intermediate strength to place the materials at the delicate boundary between itinerancy and localization. Our description takes into account the interplay between the itinerant and local-moment aspects, which are naturally associated with the interaction-induced coherent and incoherent parts of the electronic excitations. Enhancement of the spectral weight associated with the coherent electronic excitations weakens the magnetic order, and induces a magnetic quantum critical point. Our characterization of the magnetic excitations is important not only for the understanding of the existing and future experiments in the normal state, but also for the microscopic understanding of high-temperature superconductivity in the iron pnictides and related metallic systems close to a Mott transition. In addition, realization of a magnetic quantum critical point in the iron pnictides provides a new setting to explore some of the rich complexities (42, 43) of quantum criticality; this is much needed since quantum critical points have so far been explicitly observed only in a very small number of materials.

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