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Theory of the bound state of 4*f* excitations and magnetic resonance in unconventional superconductors

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We analyze the influence of unconventional superconductivity on crystalline electric-field (CEF) excitations of rare-earth ions in layered superconductors. We show that resonant magnetic excitations of the conduction electrons that have been observed in these systems below T_c may result in the formation of the bound state in the 4f-electron susceptibility at energies well below the CEF excitation energy. Our results are in agreement with the observed increase of the linewidth of CEF excitations below T_c in superconducting ferropnictides and support the $s^{\pm}$ Cooper-pairing state in these compounds.

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The influence of the crystalline electric-field (CEF) splitting of rare-earth impurities on the various thermodynamic properties in metals is well known.[1] It is also known that CEF split rare-earth impurities in a conventional s-wave superconductor influence the superconducting transition temperature quite differently than non-CEF split magnetic ions described by the Abrikosov-Gor'kov theory. In addition there is also a feedback of conduction electrons on the CEF splitting.[2] In conventional s-wave superconductors the Landau damping is strongly reduced for energies smaller than twice the superconducting gap, $2\Delta_0$. In case when the excitation energy between the ground and the first-excited CEF states Δ_{CEF} is comparable to $2\Delta_0$ one finds line narrowing of the lowest CEF excitations and a slight shift in frequency toward higher energies when the temperature T falls below T_c. This is a simple consequence of the fact that both real and imaginary parts of the conducting electron-spin response are suppressed in an s-wave superconducting state for frequencies $\omega \leq 2\Delta_0$.

The situation is different in unconventional high-T_c superconductors such as the layered cuprates or ferropnictides which may contain rare-earth elements. In these systems the superconducting gaps are often comparable and even larger than Δ_{CEF}. This allows to study in detail the feedback effect of superconductivity on CEF excitations. For example, some attempts have been made to analyze the symmetry of the superconducting gap in layered cuprates by looking at the CEF linewidth as a function of temperature with $\Delta_{CEF} \ll 2\Delta_0$.[3] A much more dramatic effect on CEF excitations is the strong change in the conduction-electron-spin susceptibility below T_c and its consequence for the 4f response. In unconventional superconductors this change often results in a resonance peak which has been observed by inelastic neutron scattering (INS) (Refs. 4–6) near the antiferromagnetic wave vector $\mathbf{Q}$ in a number of systems like high-T_c cuprates,[4] UPd_2Al_3,[7] $CeCoIn_5$,[8] $CeCu_2Si_2$,[9] and recently also in ferropnictides.[5] It depends sensitively on the type of unconventional Cooper pairing and may also be used to eliminate certain forms of pairing.[10]

Interestingly, the rare-earth elements with unfilled 4f shells in $ReFeAsO_{1-x}F_x$ (Re=Sm,Ce,Gd,Pr) systems result in a number of peculiar properties as compared to the La-based counterparts. For example, in addition to the known enhancement of the superconducting transition temperature, one also finds an increase of the magnetic moments on the Fe sites in the spin-density wave (SDW) phase[11] as well as a non-Fermi-liquid temperature dependence of the resistivity.[12] Altogether these facts point toward larger influence of the rare-earth ions on the electronic properties of the Fe_2As_2 layers as it was originally anticipated. Moreover, recent neutron-scattering studies reported anomalous broadening of the CEF excitations below the superconducting transition temperature in the $CeFeAsO_{0.84}F_{0.16}$,[13] which arises due to the coupling to the conduction electrons.

In this Rapid Communication we analyze the effect of a feedback resonance below the superconducting transition temperature on CEF excitations of the 4f states. Depending on the strength of the coupling between the 4f and d electrons, the resonance causes the formation of the bound state (an additional pole) in the 4f-electron susceptibility for electron- and hole-doped cuprates. We further address the anomalous features of the CEF excitations found in the $CeFeAsO_{0.84}F_{0.16}$ superconductor.[13,23] In particular, we show that an increase of the CEF linewidth below T_c observed experimentally can be explained *only* under assumption that the gap function has different signs on the electron- and hole-Fermi surfaces and is fully consistent with $s^{\pm}$ Cooper pairing.

The presence of rare-earth ions with 4f electrons in a metallic matrix with d electrons forming the conduction band can be described by the model Hamiltonian

$$H = \sum_{i\gamma} \varepsilon_\gamma |i\gamma\rangle\langle i\gamma| + \sum_{\mathbf{k},\sigma} \varepsilon_{\mathbf{k}} d^\dagger_{\mathbf{k}\sigma} d_{\mathbf{k}\sigma} + U\sum_{i,m} n_{i\uparrow} n_{i\downarrow}. \quad (1)$$

Here, $|i\gamma\rangle$ denotes a CEF eigenstate γ of the incomplete 4f-shell at lattice site i, and $d^\dagger_{\mathbf{k}\sigma}$ creates an electron in the conduction band with wave vector $\mathbf{k}$ and spin σ. The ener-

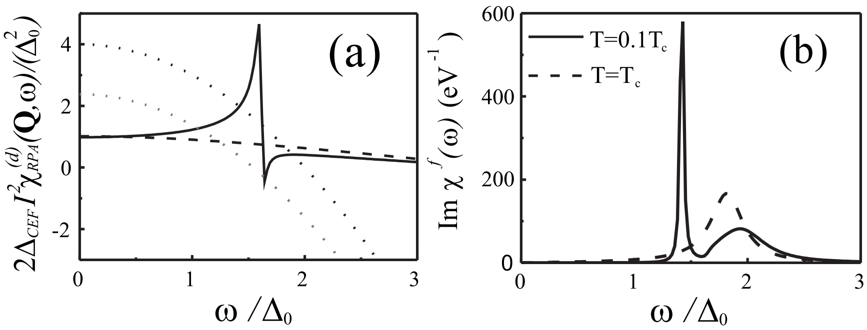

FIG. 1. (a) Graphical illustration of the solution of Eq. (4). The dotted curve refers to the function $\Delta_{CEF}^2-\omega^2$ with $\Delta_{CEF}=2\Delta_0$ (black) and $\Delta_{CEF}=1.5\Delta_0$ (gray), while $2\Delta_{CEF}I_0^2\chi_{RPA}^{(d)}(\mathbf{q},\omega_{\mathbf{q}})$ is shown for the normal (dashed curve) and the superconducting (solid curve) state assuming $I_0\approx\Delta_0$. (b) Calculated Im $\chi^f(\mathbf{Q},\omega)$ at $T=T_c$ (dashed curve) and in the superconducting state ($T=0.1T_c$) (solid curve). The dispersion parameters and Coulomb repulsion for the electron gas on a square lattice have been used (Ref. 15).

gies of the CEF eigenstates are denoted by ε_γ. Furthermore, $\varepsilon_{\mathbf{k}}$ is the dispersion of the conduction band. Correlations of the d-electrons are due to an on-site electron repulsion U. The coupling term between the conduction and $4f$ electrons at site i is given by

$$H_I = -I_{ex}(g_J-1)\sum_i \mathbf{s}_i\mathbf{J}_i = -I_0\sum_i \mathbf{s}_i\mathbf{J}_i, \tag{2}$$

i.e., by the exchange interaction between itinerant d spins ($\mathbf{s}_i$) and localized $4f$ electrons determined by Hund's rule with a total angular momentum, $\mathbf{J}$. The behavior of the conduction-electron susceptibility in the superconducting state is treated within the random-phase approximation (RPA), $\chi_{RPA}^{(d)}(\mathbf{q},\omega) = \frac{\chi_0^{(d)}(\mathbf{q},\omega)}{1-U\chi_0^{(d)}(\mathbf{q},\omega)}$, where $\chi_0^{(d)}(\mathbf{q},\omega)$ is the non- interacting electron susceptibility in the superconducting state.[14] For large momenta $\mathbf{q}$ and low frequencies, Im $\chi_0^{(d)}(\mathbf{q},\omega)$ is zero and it exhibits a discontinuous jump at the onset frequency of the particle-hole continuum $\Omega_c=\min(|\Delta_{\mathbf{k}}|+|\Delta_{\mathbf{k+q}}|)$ where both $\mathbf{k}$ and $\mathbf{k+q}$ lie on the Fermi surface. Note, however, that the discontinuity in Im $\chi_0^{(d)}$ occurs only if $\mathrm{sgn}(\Delta_{\mathbf{k}})=-\mathrm{sgn}(\Delta_{\mathbf{k+q}})$ which is not possible for an isotropic s-wave order parameter. A discontinuity in Im $\chi_0^{(d)}$ leads to a logarithmic singularity in Re $\chi_0^{(d)}$. As a result, the resonance conditions (i) U Re $\chi_0^{(d)}(\mathbf{q},\omega_r)=1$ and (ii) Im $\chi_0^{(d)}(\mathbf{q},\omega_r)=0$ can both be fulfilled at $\omega_r<\Omega_c$ for any value of $U>0$. This results in a resonance peak below T_c in form of a spin exciton. For finite quasiparticle damping Γ, condition (i) can only be satisfied if $U>0$ exceeds a critical value, while condition (ii) is replaced by U Im $\chi_0^{(3d)}(\mathbf{q},\omega_r)\ll 1$.

Typically a CEF splits the Hund's rule $\mathbf{J}$-multiplet of the incomplete $4f$-shell with different CEF levels. For Ce^{3+} ions these levels are either three Kramers doublets or a doublet and a quartet, depending on the symmetry of the CEF. We assume for simplicity a two-level system (TLS) only consisting of two doublets. The splitting is Δ_{CEF} and the susceptibility of this TLS is $u_\alpha(\omega)=|m_\alpha|^2\frac{2\Delta_{CEF}}{(\Delta_{CEF}^2-\omega^2)}$ with $|m_\alpha|^2 = \Sigma_{ij}|\langle i|J_\alpha|j\rangle|^2\tanh(\beta\Delta_{CEF}/2)$. For the sake of simplicity we further assume $|m_z|\ll|m_x|$, and $|m_x|=|m_y|=|m_\perp| = m_0\tanh(\beta\Delta_{CEF}/2)$ and set $m_0=1$.

The $4f$-electron susceptibility within RPA approximation is given by

$$\chi^f(\mathbf{q},\omega) = \frac{u_\alpha(\mathbf{q},\omega)}{1-I_0^2 u_\alpha(\mathbf{q},\omega)\chi_{RPA}^{(d)}(\mathbf{q},\omega)}. \tag{3}$$

The position of the pole and its damping by the imaginary part of χ^f can be probed by INS. It is determined by

$$\Delta_{CEF}^2-\omega_{\mathbf{q}}^2-2\Delta_{CEF}I_0^2|m_\perp|^2[\chi_{RPA}^{(d)}(\mathbf{q},\omega)]'=0,$$

$$\Gamma_q = 2I_0^2\Delta_{CEF}|m_\perp|^2[\chi_{RPA}^{(d)}(\mathbf{q},\omega_{\mathbf{q}})]''. \tag{4}$$

In the normal state Re $\chi_{RPA}^{(d)}(\mathbf{Q}_{AF},\omega)$ is a positive function, which decreases with increasing frequency. As a consequence the actual position of the peak in Im χ^f will be shifted toward lower frequency and acquire an additional damping that originates from the Landau damping in the case of Im $\chi_0^{(d)}$, or as in the case of Im $\chi_{RPA}^{(d)}$ by the interaction with the overdamped spin waves of the itinerant electrons. This is shown in Fig. 1(a) where we present the numerical solution of Eq. (4) for the normal state. The actual position and the shift depend on the strength of the coupling of the conduction electrons to the $4f$ electrons.

In the superconducting state with unconventional order parameter such that $\Delta_{\mathbf{k}}=-\Delta_{\mathbf{k+Q}}$, the conduction-electron susceptibility has a resonance peak at energy $\omega_r\leq|\Delta_{\mathbf{k}}|+|\Delta_{\mathbf{k+Q}}|$. Observe that Re $\chi_{RPA}^{(d)}(\mathbf{Q},\omega)$ is discontinuous at ω_r and changes sign from positive at $\omega<\omega_r$ to the negative at $\omega>\omega_r$ depending on the strength of the interaction between $4f$ and $3d$ electron, the occurrence of the resonance in the d-electron susceptibility yields interesting consequences for the $4f$ electron susceptibility.

In the strong-interaction regime, determined by condition $I_0^2\geq\frac{\Delta_{CEF}^2-\omega_r^2}{\Delta_{CEF}\,\mathrm{Re}\,\chi_{RPA}^{(d)}(\mathbf{Q},\omega_r)}$, the $4f$-electron susceptibility will have an additional pole (bound state[16]) at energies well below Δ_{CEF}. Simultaneously, due to the change of sign of the $\chi_{RPA}^{(d)}(\mathbf{q},\omega)$ for $\omega>\omega_r$, the initial position of the Δ_{CEF} will be shifted to higher frequencies. The resulting pole structure and Im $\chi^f(\mathbf{Q},\omega)$ are shown in Fig. 1. Note that, due to the interaction between the $4f$ shell and d electrons and assuming $\Delta_{CEF}\simeq 2\Delta_0$, the positions of the new poles are approximately given by $\omega_q^{\pm}\simeq\frac{1}{2}(\Delta_{CEF}+\omega_r)\pm\frac{1}{2}\sqrt{(\Delta_{CEF}-\omega_r)^2+4I_0^2|m_\perp|^2 z_r}$ where z_r is residue of the resonance peak in the d spin susceptibility.

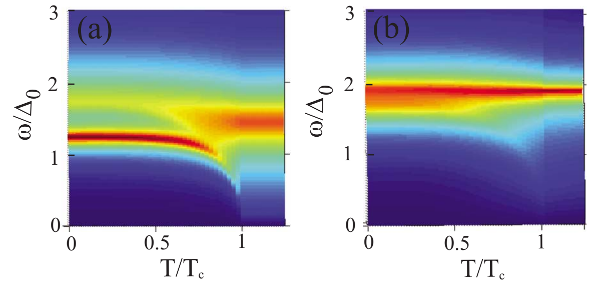

FIG. 2. (Color online) Calculated temperature dependence of the lowest CEF excitation in an unconventional superconductor for (a) strong coupling ($I_0 \approx \Delta_0$) and (b) weak coupling ($I_0 < \Delta_0$) coupling between conduction electrons and localized $4f$ electrons. The tight-binding parameters for the electron-doped cuprates (Ref. 15) and ferropnictides (Ref. 17) are used. In both cases we assume a temperature dependence of the superconducting order parameter of the form $\Delta_{\mathbf{k}}(T) = \Delta_{\mathbf{k}} \tanh(1.76\sqrt{\frac{T_c}{T}-1})$.

In Fig. 1(b) we show the resulting structure of the imaginary part of the f-electron-spin susceptibility as function of frequency at the antiferromagnetic wave vector, $\mathbf{Q}$. Due to appearance of the resonance mode in the conduction-electron susceptibility the CEF excitations acquire an additional pole below the superconducting transition temperature at energies well below $2\Delta_0$ with a small linewidth. In addition a second pole is found at a frequency which is higher than the renormalized Δ_{CEF} in the normal state. Its damping induced by Im $\chi^{(d)}_{RPA}$, however is still larger than it is in the normal-state value as a consequence of discontinuous jump of Imχ_0 at $2\Delta_0$. Therefore, CEF excitations behave completely opposite as in conventional superconductors with $\Delta_{CEF} < 2\Delta_0$. Notice that in case Δ_{CEF} becomes smaller than $2\Delta_0$ the position of the bound state shifts to lower energies [see Fig. 1(a)], and for $\Delta_{CEF} < 1.5\Delta_0$ one finds only a single pole as a result of the coupling between d and $4f$-susceptibilities. In Fig. 2(a) we show the temperature evolution of the CEF excitations below the superconducting transition temperature. Close to T_c, i.e., in the range $0.8T_c < T < T_c$, the splitting of the CEF excitations due to formation of the magnetic resonance is not well resolved and it looks as if the CEF level gets damped anomalously strong. With lowering temperature, i.e., $T \approx 0.8T_c$, both peaks become well separated and the lower mode is much narrower.

In the weak-coupling case, i.e., when the coupling between conduction electrons and localized $4f$ electrons is small, the effect of unconventional superconductivity on the CEF excitations still differs substantially from that of conventional superconductors. In particular, for $I_0 < \Delta_0$ there is no additional peak in Im χ^f below T_c, which is due to relatively small value of $I_0^2\chi^{(d)}_{RPA}(\mathbf{q},\omega_{\mathbf{q}})$. In other words, even though $\chi^{(d)}$ is strongly frequency dependent at low energies the coupling is not large enough to produce a second pole in the f-electron susceptibility. Nonetheless, the effect of the resonance in the former is present in this frequency range. Because the overall conduction-electron response of the unconventional superconductor is larger than its normal-state counterpart, the CEF excitations below T_c will experience larger damping due to Im $\chi^{(d)}_{RPA}(\mathbf{Q},\omega \sim 2\Delta_0)$ which is enhanced in the superconducting state. Similarly Re $\chi^{(d)}_{RPA}(\mathbf{Q},\omega \sim 2\Delta_0)$ is lower than its normal-state value but, more important, it changes sign above ω_r [see Fig. 1(a)]. As a result the CEF excitation simultaneously requires larger damping and also shifts toward higher frequencies. We show the evolution of the CEF excitation in Fig. 2(b) as a function of temperature where below T_c this effect is clearly visible.

The description above is correct under the assumption that there is a well-defined resonant excitation in the conduction-electron susceptibility at the antiferromagnetic wave vector, $\mathbf{Q}$. This condition is realized particularly well in quasi-two-dimensional systems such as hole- and electron-doped cuprates[15] as well as in the iron-based superconductors[17] and also in other heavy-fermion systems such as $CeCoIn_5$.[10]

In the following we show that recently observed shift on anomalous damping of the CEF excitation in $CeFeAsO_{1-x}F_x$ is a consequence of $s^{\pm}$ superconducting order.[13,23] In particular, as discussed above if the coupling between the conduction electrons and Ce ions is relatively weak ($I_0 < \Delta_0$) the effect of unconventional superconductivity on the CEF excitations does not result in the appearance of an additional pole in Im χ_f. But, because of the sign change of the superconducting gap for the electron and the hole Fermi surfaces connected by $\mathbf{Q}_{AF}$, Im $\chi^{(d)}_{RPA}$ is enhanced in the superconducting state when $\omega > \omega_r$. Simultaneously, the real part of χ_f changes sign and becomes negative for $\omega > \omega_r$, i.e., Δ_{CEF} shifts to higher energies below T_c [see Fig. 2(b)]. To compare the results with experiments, we adopt the parameters used previously for calculations of the itinerant magnetic excitations in ferropnictides[17] and assume the coupling of the itinerant $3d$ electrons to the $4f$-shell $I_0 \simeq 0.72\Delta_0 < \Delta_0$. We find that in the superconducting state, due to the $s^{\pm}$ symmetry of the gap function the RPA spin susceptibility between electron and hole pockets centered around the Γ and M points of the BZ, respectively, shows the characteristic enhancement at energies less than $2\Delta_0$.[17,18] Due to weak coupling to the $4f$ shell this does not yield a second pole in Imχ_f but instead results in an anomalous damping of the CEF excitations and a shift of the characteristic frequency, ω_q, toward higher energies as shown in Fig. 3. This is in striking agreement with the experimental data on $CeFeAsO_{0.84}F_{0.16}$.[13]

Other systems where similar effects can occur are layered cuprates with rare-earth ions. For example, in the electron-doped cuprate system $Nd_{2-x}Ce_xCuO_4$, the splitting between the ground and the first CEF excited levels of the $4f$ multiplet for Nd^{3+} ions is about $\Delta_{CEF} \approx 20$ meV,[19] which is comparable to the energy of $2\Delta_0$ of the superconducting gap.[15] The situation with the position of the feedback spin resonance due to the $d_{x^2-y^2}$-wave symmetry of the superconducting gap in the electron-doped cuprates is far from being well understood. For example, originally the resonance had been reported in $Pr_{0.88}LaCe_{0.12}CuO_{4-\delta}$ and $Nd_{1.85}Ce_{0.15}CuO_4$ at energies of about 12 and 10 meV, respectively.[20,21] Recently, this conclusion has been challenged by another group[22] where the feedback spin exciton in $Nd_{1.85}Ce_{0.15}CuO_4$ has been found at much smaller energies around 4.5 and 6.4 meV. In Fig. 1(b) we show results for Im χ_f assuming a tight-binding energy dispersion for the $3d$-electrons,[15] a coupling-constant $I_0 = \Delta_0$ and $\Delta_{CEF} = 2\Delta_0$. Here the f-electron susceptibility shows two peaks below T_c with the lower one

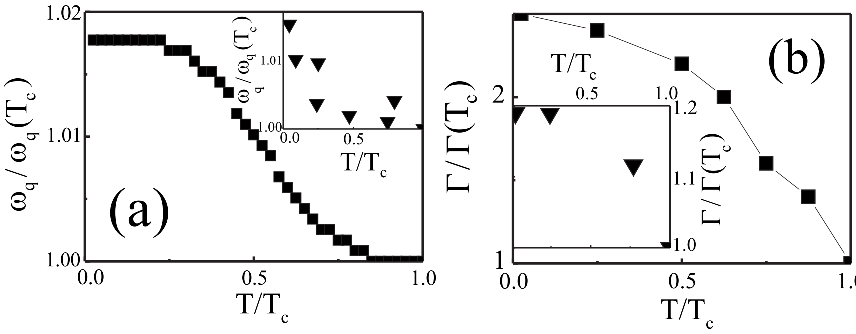

FIG. 3. Calculated temperature dependence of the frequency position, ω_q (a) and the linewidth (b) of the lowest CEF level assuming weak coupling between the conduction and the 4f-shell. The insets show the corresponding experimental data for the superconducting CeFeAsOF system (Ref. 13).

at the energy smaller than ω_r and the upper one slightly above the renormalized Δ_{CEF} in the normal state. In order to see whether the lower peak found recently[22] is indeed related to the bound state discussed here further studies are necessary. A hallmark of this would be the large contribution of the incomplete 4f shell to the d electron susceptibility spin resonance. In addition, below T_c the CEF excitations should possess larger damping in the superconducting state than in the normal state and show slight shift toward higher energies.

In conclusion, we find that the feedback of unconventional superconductivity on the CEF excitations results in two characteristic features. If the coupling between the CEF excitations and conduction d-electrons is large ($I_0 \sim \Delta_0$), the resonant excitations in the conducting electrons susceptibility centered at ω_r yield an additional bound state in the f-electron susceptibility at energies $\omega \leq \omega_r$. At the same time, the CEF excitations shifts toward higher energies and acquire an additional damping below T_c. If the coupling between the d electrons and CEF excitations is weak, i.e., $I_0 < \Delta_0$ the additional pole does not occur and the only effect of the unconventional superconductivity is the anomalous damping of the CEF excitations and their slight upward shift below T_c. We have shown that the latter effect is observed in iron-based superconductors and supports strongly the $s^{\pm}$ gap function which changes sign between electron and hole pockets.

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