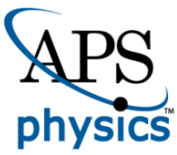

Pairing Mechanism in Hund's Metal Superconductors and the Universality of the Superconducting Gap to Critical Temperature Ratio

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On the Pairing Mechanism in Hund's Metal Superconductors and the Universality of the Superconducting Gap to Critical Temperature Ratio

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We analyze a simple model containing the physical ingredients of a Hund's metal, the local spin fluctuations with power-law correlators, $(\Omega_0/|\Omega|)^\gamma$, with γ greater than one, interacting with electronic quasiparticles. While the critical temperature and the gap change significantly with varying parameters, the $2\Delta_{max}/k_BT_c$ remains close to twice the BCS value in agreement with experimental observations in the iron-based superconductors (FeSC).

Introduction The discovery of superconductivity in iron-based materials [1] opened a new area of research in the field of superconducting materials. There are by now many families of materials which are based on tetrahedrally coordinated irons to pnictides and chalcogenides, with different separating layers in between. For a review, see Refs. 2 and 3. Photoemission studies have shown that some of these compounds have bands which are well described by the standard density functional theory with small renormalizations, while in others, the mass renormalizations are larger than ten. Hence, it is agreed upon that in these class of compounds the strength of the correlation varies substantially. On the other hand, there is no agreement on the type of correlation, which is attributed to Mott [4] or Hund's physics [5–7].

Thinking of the iron pnictides as Hund's metals presents a scenario, in which the physics governing the behavior of different materials is the same, but the correlations are sensitive to the filling of the shell and the height of the pnictogen/chalcogen ligand [8]. A deeper understanding of Hund's metal physics shows that the normal state above the superconducting transition has a broad intermediate region of temperatures characterized by orbital spin separation, whereby the spin excitations are quasi-atomic like, while the orbital excitations are fully itinerant [9–11]. Thus, the Hund's metal behavior exists in a temperature range below the Kondo scale of the orbital degrees of freedom, T_K^{orb}, and above the Kondo scale of the spin degrees of freedom, T_K^{sp}, below which the Fermi liquid holds [11].

Whether a Hund's metal becomes a superconductor at high temperatures is expected to depend on many microscopic details such as the shape of the Fermi surfaces of the electrons, the dispersion of the spin excitations, and how they are coupled to electrons [12, 13]. Hence the superconducting critical temperature is not a universal quantity, much like a coherence-incoherence crossover where a Fermi liquid emerges from a Hund's metal state.

In this paper, we point out a universal aspect of superconductivity, which emerges from a Hund's metal state at higher temperatures – we argue that while T_c and the maximum value of the superconducting gap at $T = 0$, Δ_{max}, are material-dependent, their ratio $2\Delta_{max}/T_c$ is material-independent universal number. We show that this is the case if the pairing in a Hund metal is mediated by quasi-local spin excitations. As the normal state of a Hund's metal, involves incomplete screening, it is characterized by a power-law behavior of all the physical quantities [5, 9–11]. In particular, the susceptibility of local spin fluctuations has a power law dependence above a characteristic Kondo scale. While an analytic theory of such power-law behavior is not yet available, the numerical studies and physical considerations clearly indicate that the spin susceptibility follows $\chi(\Omega) \propto 1/|\Omega|^\gamma$ with $\gamma > 1$ [11]. Here we show that, when such $\chi(\Omega)$ mediates superconductivity emerging from the Hund's metal state, the ratio $2\Delta_{max}/k_BT_c$ is a universal, $\gamma-$dependent number, which for $\gamma > 1$ is substantially larger than the BCS value. Universality here means that this number does not depend on the strength of the coupling to magnetic fluctuations, while T_c and Δ_{max} vary strongly with the strength of fermion-boson coupling.

These results are in agreement with the conclusions of recent experiments on FeSC which addressed this question from an experimental perspective. By measuring the gap and the critical temperature in LiFeAs and $FeTe_{0.55}Se_{0.45}$, Miao *et al.* established [14] that in both systems $2\Delta_{max}/k_BT_c \sim 7.2$, despite that the electronic structures are different. The previous study on the spin-resonance also found a universal ratio, Ω_{res}/k_BT_c [15]. This last observation is consistent with the universality of $2\Delta_{max}/k_BT_c$ if Ω_{res} scales with Δ_{max} as numerous studies of spin resonance suggested [16].

Hund's metals are not confined to the iron-based superconductors and are in fact very common. Sr_2RuO_4 is a prime example of Hund's metal [17–21]. However, for Sr_2RuO_4, its T_K^{sp} is much higher than its superconducting temperature [18]. Consequently, the normal state above the superconducting transition is already a Fermi-liquid instead of a Hund's metal as in FeSCs. Therefore, our theory does not apply there.

The model To describe superconductivity in Hund's

metals we use the γ-model, which was introduced in the context of superconductivity near a quantum critical point [22–31]. Namely, we assume that interaction between fermions is mediated by a local spin susceptibility $\chi(\Omega) \propto 1/|\Omega|^\gamma$ [32]. This interaction simultaneously gives rise to pairing and frequency-dependent fermionic self-energy $\Sigma(\omega)$. The γ-model ignores many of the complications of a realistic description of the iron pnictide superconductors: a) its multiband and multiorbital nature, b) multiple Fermi surfaces, c) orbital-induced gap variation along the Fermi surfaces and the variation of the phase and magnitude of a superconducting order parameter between different Fermi surfaces, d) fine features in the dynamical structure factor of spin fluctuations (see Refs. 7, 33–37 for recent reviews). It retains, however, two essential features, the superlinear divergence of the local spin susceptibility at intermediate frequencies, and the coupling of quasi-localized spins to fermionic quasiparticles. We argue that this is the essential ingredient to understand the results of Ref. [14] that a) the ratio of $2\Delta_{max}/k_BT_c$ is much larger than in BCS theory and b) it does not vary between different materials, as opposed to T_c and Δ_{max}, which both are material-dependent.

The expressions for T_c and Δ_{max} in the γ-model are obtained by solving the set of Eliashberg equations [24, 26, 27, 31, 32, 38] for the pairing vertex $\Phi(\omega_n)$ and fermionic self-energy $\Sigma(\omega_n)$ with a power-law form of the interaction:

$$\Sigma(\omega_n) = \pi T \sum_{\omega_m} \lambda(\omega_m - \omega_n) \frac{\omega_m + \Sigma(\omega_m)}{\sqrt{(\omega_m + \Sigma(\omega_m))^2 + \Phi^2(\omega_m)}}, \tag{1}$$

$$\Phi(\omega_n) = \pi T \sum_{\omega_m} \lambda(\omega_m - \omega_n) \frac{\Phi(\omega_m)}{\sqrt{(\omega_m + \Sigma(\omega_m))^2 + \Phi^2(\omega_m)}}, \tag{2}$$

where

$$\lambda(\Omega) = \left(\frac{\Omega_0}{|\Omega|}\right)^\gamma, \tag{3}$$

and Ω_0 determines the strength of fermion-boson coupling. The two Eliashberg equations can be partly factorized by introducing the pairing gap $\Delta(\omega_n) = \Phi(\omega_n)\omega_n/(\omega_n + \Sigma(\omega_n))$ instead of $\Phi(\omega)$. With this substitution, the self-energy $\Sigma(\omega_n)$ drops from the equation for $\Delta(\omega)$. We have

$$\Delta(\omega_n) = \pi T \sum_{\omega_m} \frac{\lambda(\omega_m - \omega_n)}{\sqrt{\omega_m^2 + \Delta^2(\omega_m)}} \left(\Delta(\omega_m) - \Delta(\omega_n)\frac{\omega_m}{\omega_n}\right). \tag{4}$$

The $\lambda(\Omega)$ diverges at $\Omega = 0$, when $\omega_n = \omega_m$. However, the term in the bracket in the r.h.s of Eq. 4 becomes zero at $\omega_n = \omega_m$, which cancels out the divergence. Hence, Eq. 4 is free from singularities at any finite T. The equation on $\Sigma(\omega)$ does depend on $\Delta(\Omega_n)$:

$$\Sigma(\omega_n) = \pi T \sum_{\omega_m} \lambda(\omega_m - \omega_n) \frac{\omega_m}{\sqrt{\omega_m^2 + \Delta^2(\omega_m)}}. \tag{5}$$

Because of semi-factorization, one has to solve first Eq. 4 for $\Delta(\omega_n)$, substitute the result into Eq. 5 and obtain $\Sigma(\omega_n)$.

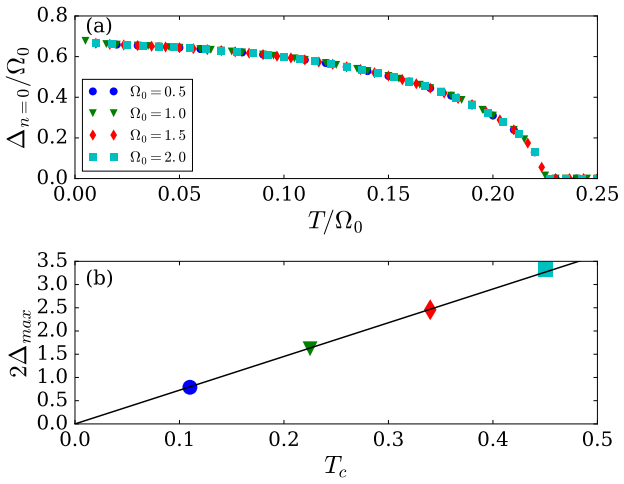

FIG. 1. (Color online) (a) The pairing gap at the first Matsubara frequency , $\Delta_{n=0}$, as a function of temperature, T, for different pairing amplitudes, Ω_0, at $\gamma = 1.2$. (b) The maximum gap, Δ_{max}, and critical temperature, T_c, for various paring amplitudes, Ω_0, at $\gamma = 1.2$. The black solid line is the linear fit to the slope, $2\Delta_{max}/T_c = 7.2$, corresponding to the experimental value observed in FeSC.

In quantum-critical theories, $\gamma = 2$ corresponds to the strong coupling limit of electron-phonon interaction [39], $\gamma = 1/2$ describes pairing by antiferromagnetic spin fluctuations in 2D[22, 24, 25], $\gamma = 1/3$ describes pairing by a gauge field and ferromagnetic spin fluctuations in 2D [30, 40–44], $\gamma = 0+$ describes color superconductivity and pairing in 3D [45, 46]. The models with varying $\gamma < 1$ have also been analyzed [26, 27, 29, 31]. Here we use the fact that in a wide range of frequencies a Hund's metal is also characterized by a local susceptibility, $\chi(\Omega) \propto 1/|\Omega|^\gamma$, with γ greater than one [32], and explore the consequences of such a model on the $2\Delta_{max}/T_c$ ratio by numerically and analytically solving Eq. 3 and Eq. 4. We obtain $\Delta(\omega_m)$ on the Matsubara axis and convert it onto real axis by analytical continuation. We define Δ_{max} at $T = 0.005\Omega_0$ as the frequency at which the density of states $N(\omega) \propto Im\big(\omega/(\Delta^2(\omega) - \omega^2)\big)$ jumps to a finite value, i.e., set $\Delta_{max} = \Delta(\omega = \Delta_{max})$.

The results Figure 1(a) shows our results of the pairing gap at the first Matsubara frequency, $\Delta_{n=0}$, as a function of temperature, T, for a given $\gamma = 1.2$. We see that $\Delta_{n=0}$, measured in units of the interaction strength Ω_0, is a universal function of T/Ω_0 (i.e., the functional form does not depend on Ω_0). This can be seen directly from

Eq. 4 by simultaneously rescaling $\Delta(\omega_n)$ and Matsubara frequencies $\omega_{n,m}$ by Ω_0. For this particular γ we obtained $\Delta_{max} = 0.69\Omega_0 \approx \Delta(\pm\pi T)$ and $T_c = 0.19\Omega_0$. The ratio $2\Delta_{max}/T_c = 7.2$ is the universal number, independent on Ω_0, as we explicitly show in Fig. 1(b). This universality is indeed the consequence of the fact that Ω_0 is the only energy scale in the problem. For a generic γ, we expect $T_c = A(\gamma)\Omega_0$ and $\Delta_{max} = B(\gamma)\Omega_0$, i.e., $2\Delta_{max}/T_c = 2B(\gamma)/A(\gamma)$.

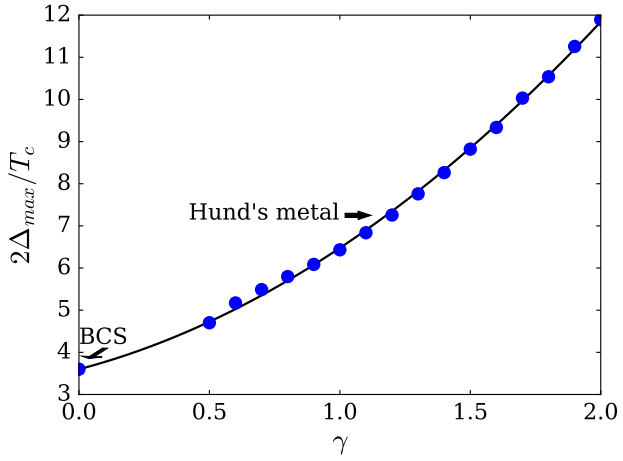

FIG. 2. (Color online) The ratio, $2\Delta_{max}/T_c$, as a function of γ. The arrows indicate the ratio $2\Delta_{max}/T_c = 7.2$ ($\gamma \sim 1.2$) and 3.6 ($\gamma = 0$) corresponding to the ratio for FeSC and BCS. The black solid line is the fit to a parabola with $2\Delta_{max}/T_c = 3.6 + 1.63\gamma + 1.24\gamma^2$.

It is instructive to study how the ratio, $2\Delta_{max}/T_c$, varies with the exponent γ because different γ describe different pairing mechanisms. We show our numerical results for $2\Delta_{max}/T_c$ for various γ in Fig. 2. The ratio, $2\Delta_{max}/T_c$, increases with increasing γ in a parabolic fashion which can be extrapolated to the BCS ratio, $2\Delta_{max}/T_c = 3.6$, at $\gamma \geq 0$. We found that the experimental $2\Delta_{max}/T_c \sim 7.2$, reported by Miao *et al.*, is reproduced for $\gamma \sim 1.2$. Remarkably, this value of γ coincides with the exponent of the local spin susceptibility, obtained from the extensive numerical analysis of Hund's metal state in the three-band Hubbard model [9, 11]. This agreement is the strong argument that incoherent spin fluctuations, specific to a Hund's metal state, may indeed mediate superconductivity in FeSCs.

To get further insight into this issue, we now discuss how Δ_{max}, T_c, and also fermionic $\Sigma(\omega_m)$ individually vary with γ. To get $T_c(\gamma)$ and self-energy near T_c we follow [26, 27] and (a) solve for T at which the linearized gap equation (the one with infinitesimally small $\Delta(\omega_m)$) has the solution, and (b) solve Eq. 5 at $\Delta_n = 0$.

We show the result of numerical calculation of $\Sigma(\omega_n)$ in Fig. 3(a). Analytical reasoning shows [26] that, at large ω_n, $\Sigma(\omega_m)$ scales as $\omega^{1-\gamma}$ for $0 < \gamma < 1$ and saturates to $\Sigma(\omega_m) = (\Omega_0^\gamma/(2\pi T)^{\gamma-1})\zeta(\gamma)$ for $\gamma > 1$, where $\zeta(\gamma)$ is the Riemann zeta-function. Our numerical results fully reproduce this asymptotic behavior. In Fig. 3(b) we show the numerical result for the prefactor $A(\gamma)$ in the critical temperature, $T_c = A(\gamma)\Omega_0$. The analytical expression for $A(\gamma)$ has been obtained in Ref. 27 within large N approximation. An extension of that result to the physical case $N = 1$ yields $A(\gamma) = \frac{1}{2\pi}\left(1+\frac{\delta_\gamma}{\gamma}\right)$, where δ_γ is a number in the order of one ($\delta_{\gamma\gg1} \approx 1/2$). Our numerical result is consistent with this formula, particularly the increase of

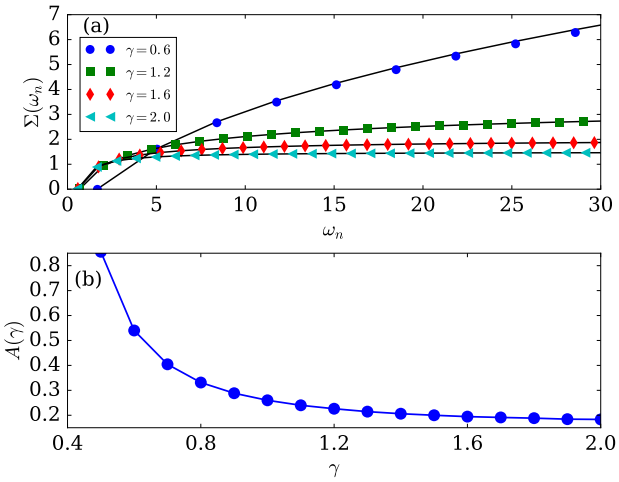

FIG. 3. (Color online) (a) The Matsubara self-energy, $\Sigma(\omega_n)$, for different power, γ, with $\Omega_0 = 1$ near critical temperature, T_c. The black solid line is the analytic solution, from Moon *et al.* [26]. (b) The coefficient, $A(\gamma)$, corresponding to the critical temperature, $T_c = A(\gamma)\Omega_0$, as a function of power γ.

$A(\gamma)$ at smaller γ and the saturation of $A(\gamma)$ at $1/2\pi$ at larger γ (for $\gamma = 2$ we found $A(2) = 0.18$, in good agreement with Ref. 27).

At low temperatures, the pairing gap $\Delta(\omega_n)$, is no longer a small quantity. The linearization trick is no longer applicable, and we have to solve the full non-linear gap equation for $\Delta(\omega_n)$ and convert the result to the real frequency axis. In Fig. 4 we show the results for the self-energy $\Sigma(\omega_n)$ and the prefactor $B(\gamma)$ in $\Delta_{max} = B(\gamma)\Omega_0$, obtained from Eq. 5 using the solution of $\Delta(\omega_n)$, for $T = 0.005\Omega_0$. The self-energy $\Sigma(\omega)$ (Fig. 4(a)) scales linearly with ω_n at small frequencies, as expected in a Fermi liquid. The restoration of Fermi liquid behavior is the known feedback effect from superconductivity, which, e.g., accounts for peak-dip-hump behavior in cuprate superconductors below T_c (see, e.g., Ref. [47]). In physical terms, this happens because a finite gap reduces quasiparticle scattering at low frequencies and makes low-energy states longer lived. The slope of $\Sigma(\omega_n)$ at low frequency increases with increasing γ, indicating that correlations get stronger.

The behavior of $B(\gamma)$ is shown in Fig. 4(b). At small γ, $B(\gamma)$ decreases rather abruptly with increasing γ. At

larger γ, $B(\gamma)$ passes through a minimum at $\gamma \sim 1.2$ and slowly increases for $\gamma > 1.2$. The ratio $2\Delta_{max}/T_c$ (Fig. 2) is determined by the ratio between $B(\gamma)$ (Fig. 4(b)) and $A(\gamma)$ (Fig. 3(b)). At small γ, both $A(\gamma)$ and $B(\gamma)$ strongly evolve with γ, but the decrease of $B(\gamma)$ with increasing γ roughly follows the trend of $A(\gamma)$. As a result, the ratio $2B(\gamma)/A(\gamma)$ increases with increasing γ but varies not as strongly as $A(\gamma)$ and $B(\gamma)$. For larger $\gamma > 1.2$, $A(\gamma)$ saturates and the enhancement of $2\Delta_{max}/T_c$ is due to the increase of $B(\gamma)$.

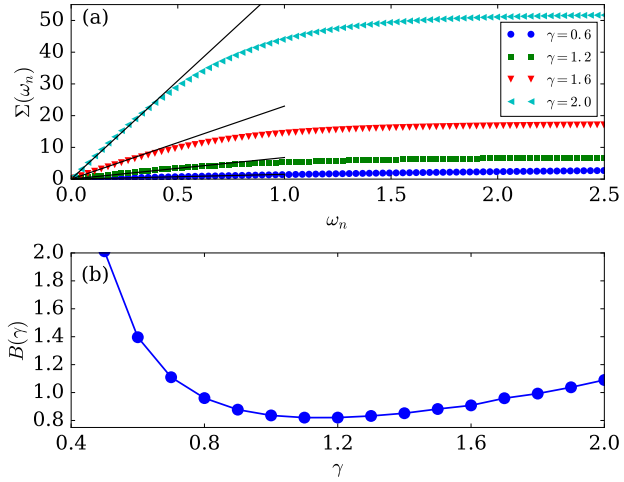

FIG. 4. (Color online) (a) The Matsubara self-energy, $\Sigma(\omega_n)$ for different power, γ, at $\Omega_0 = 1$ and $T = 0.005\Omega_0$. The solid black lines indicate the low-frequency linear behavior. (b) The coefficient, $B(\gamma)$, corresponding to the maximum of the pairing gap, $\Delta_{max} = B(\gamma)\Omega_0$, as a function of the exponent γ.

Discussion Motivated by the theoretical understanding we reexamined $2\Delta_{max}/T_c$ in other Fe-based superconductors. The data are summarized in Fig. 5. We see that they fall onto a single curve with the same slope $2\Delta_{max}/T_c = 7.2 \pm 1$ as in LiFeAs and $FeTe_{0.55}Se_{0.45}$. This universality is the strong argument in favor of Hund's metal description with electronic (spin fluctuation) pairing mechanism. Remarkably, the data for FeSe monolayers fall on a different curve with a smaller $2\Delta_{max}/T_c = 4 \pm 0.5$. This is consistent with the idea that in these systems the pairing may be mediated by electron-phonon interaction [48–51].

FeSCs are members of a broad class of unconventional superconductors, which also include copper oxides, heavy fermion metals, and the organic charge-transfer salts. The normal state of all these superconductors satisfies the criterion of bad metals, superconductivity appears near an antiferromagnetic phase, and T_c is a sizable fraction of the bandwidth. In this respect, FeSCs are often compared to the cuprates [52], because the bandwidths are comparable. At a face value, $2\Delta_{max}/T_c$ in underdoped cuprates is larger. However, one needs to take into account four additional considerations. First, the d-wave character of superconductivity in the cuprates modifies the $2\Delta_{\max}/T_c$ already in BCS limit (Ref. [53]). Second, superconductivity in underdoped cuprates emerges from a pseudogap regime, and Δ_{max} (the gap in the antinodal region) develops at an energy scale $T^* > T_c$. It would then be more appropriate to relate it to T^* rather than to T_c. Third, even above optimal doping, when pseudogap effects are relatively weak, phase fluctuations are not negligible, and the onset temperature T_p for the emergence of the bound pairs is larger than T_c. Eliashberg theory neglects phase fluctuations and, within it, one can only get $2\Delta/T_p$ ratio.

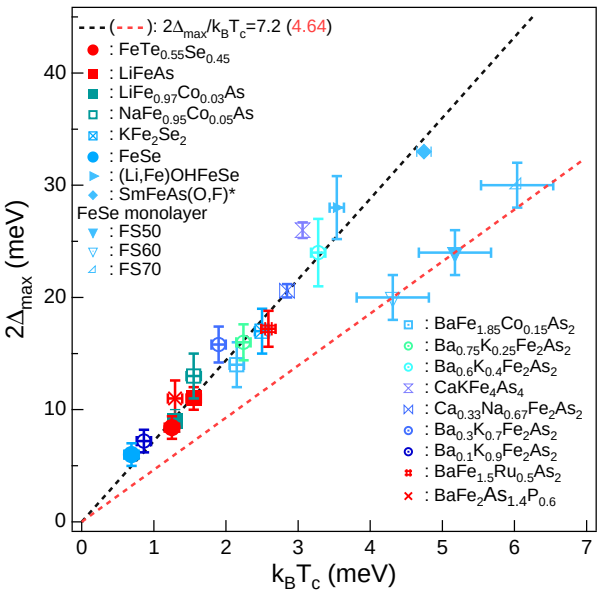

FIG. 5. (Color online) Summary of $2\Delta_{SC}^{max}/k_BT_c$ that is determined by ARPES in various bulk FeSCs [54–67] and monolayer FeSe films under different annealing conditions [68, 69]. The black and red dashed lines are linear function fit of the bulk FeSCs and monolayer FeSe, respectively. Since SmFeAs(O,F) has a non-neutral cleaved surface, the value of $2\Delta_{SC}^{max}/k_BT_c$ is extracted from the bulk sensitive optical conductivity measurement[70]. Systematic errors due to the finite instrumental resolution and the profile of the superconducting peak give 10∼17 % uncertainty of the $2\Delta_{max}/k_BT_c$ values.

Forth, there are substantial inhomogeneities in the sample, and one should compare a local temperature T_p and a local gap Δ_{max} in a given region [71] This has been done in the tunnelling studies [72], which reported $2\Delta_{max}/T_p \sim 7.9$, not that far from FeSCs. If we take all this into consideration, it appears that FeSCs and the cuprates are closer than one would have expected at first sight.

The heavy fermion superconductors also have similar $2\Delta_{max}/T_c$ ratios. For example, UPd_2Al_3 has $2\Delta_{max}/T_c = 6$ [73] and $PuCoGa_5$ has $2\Delta_{max}/T_c = 6.4 \pm 0.4$ [74], which give $\gamma \sim 1.0$. Notice that in these systems the Hund's coupling is important. On the other hand, the organic charge-transfer superconductors have ratios $2\Delta_{max}/T_c = 4.8$ corresponding to $\gamma \sim 0.5$ [75],

which are believed to be Mott systems and the Hund's physics is not relevant.

Finally, we comment on earlier realistic calculations of superconductivity in FeSCs. Yin *et al.* investigated possible pairing states using the LDA+DMFT effective pairing interaction which describes the observed spectra[76]. Due to the computational cost, they could not go to low enough temperatures to study $2\Delta_{max}/T_c$ and/or carry out an Eliashberg treatment. Nourafkan *et al.* solved the Eliashberg equations but replaced frequency-dependent interaction by a constant [77]. Ummarino carried out an Eliashberg treatment to FeScs and yields similar $2\Delta_{max}/T_c$ ratios as ours [78]. In his work, the pairing interaction was introduced phenomenologically. Combining the realistic pairing interaction with the Eliashberg approach is an outstanding challenge for future work.

Conclusions In this work, we build on the recent understanding of the physics of the Hund's metal and studied a phenomenological γ-model describing the superconductivity mediated by bosonic propagator with a power-law frequency dependence. $\lambda(\Omega) \propto 1/|\Omega|^{\gamma}$. This model captures the essence of the transition from a Hund's metal to a superconductor at a temperature comparable to or higher than a crossover temperature between non-Fermi liquid and Fermi-liquid behavior [9, 11].

We use the model to explore the main characteristics of the pairing gap and T_c, ignoring the complications such as the multiorbital/multiband structure of FeSCs. We find $2\Delta_{max}/T_c$ to be independent on the interaction strength and equal to $7.2-7.3$ if we use $\gamma = 1.2$ obtained from the three-band Hubbard model. These results are in surprisingly good agreement with recent experiments which argued that $2\Delta_{max}/T_c \approx 7.2$ is the same in at least two FeSCs: LiFeAs and $FeTe_{0.55}Se_{0.45}$ [14]. It would be interesting to extend these observations to a more realistic description of the materials, taking into account the multiorbital nature of the problem, and the fact that, in Hund's metals, the power law behavior of local spin susceptibility holds in an intermediate temperature range between a Fermi liquid regime at low temperatures and a high temperature regime where the orbitals and the spins are both quasi-atomic-like.

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