Approaching itinerant magnetic quantum criticality through a Hund’s coupling induced electronic crossover in the YFe$_2$Ge$_2$ superconductor


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Here, by conducting a systematic $^{89}$Y NMR study, we explore the nature of the magnetic ground state in a newly discovered iron-based superconductor YFe$_2$Ge$_2$. An incoherent-to-coherent crossover due to the Hund’s coupling induced electronic correlation is revealed below the crossover temperature $T^*\sim75\pm15$ K. During the electronic crossover, both the Knight shift ($K$) and the bulk magnetic susceptibility ($\chi$) exhibit a similar nonmonotonic temperature dependence, and a so-called Knight shift anomaly is also revealed by a careful $K$-$\chi$ analysis. Such an electronic crossover has been also observed in heavily hole-doped pnictide superconductors AFe$_2$As$_2$ ($A=K$, Rb, and Cs), which is ascribed to the Hund’s coupling induced electronic correlation. Below $T^*$, the spin-lattice relaxation rate divided by temperature ($1/T_1T$) shows a similar suppression as the Knight shift, suggesting the absence of critical spin fluctuations. This seems to be in conflict with a predicted magnetic quantum critical point (QCP) near this system. However, considering a $q$-dependent “filter” effect on the transferred hyperfine field, a predominant spin fluctuation with A-type correlation would be perfectly filtered out at $^{89}$Y sites, which is consistent with the recent inelastic neutron scattering results. Therefore, our results confirm that, through a Hund’s coupling induced electronic crossover, the magnetic ground state of YFe$_2$Ge$_2$ becomes close to an itinerant magnetic QCP with A-type spin fluctuations. In addition, the possible superconducting pairing due to spin fluctuations is also discussed.

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I. INTRODUCTION

Superconductivity nearby spin order is always believed to have an unconventional pairing mechanism beyond electron-phonon interactions, such as cuprate superconductors, heavy-fermion superconductors, and iron-based superconductors (FeSCs) [1–4]. Spin fluctuation is a popular candidate for gluing electrons into Cooper pairs [5]. Usually, antiferromagnetic (AFM) spin fluctuations favor spin-singlet pairing and ferromagnetic (FM) spin fluctuations favor spin-triplet pairing. In FeSCs, the stripe-type AFM spin fluctuations have been widely observed [2], which promotes early theory with spin-singlet pairing. Recently, an indirect evidence for the coexistence of AFM and FM spin fluctuations was revealed by nuclear magnetic resonance (NMR) experiments in FeSCs with 122 structure [6–8]. It is found that the FM spin fluctuations are strongest in the maximally electron- and hole-doped BaCo$_2$As$_2$ and KFe$_2$As$_2$ [7]. This strongly suggests that the competition between AFM and FM spin fluctuations is a crucial ingredient to understand the variability of superconducting temperature ($T_c$) [7], especially for the domelike behavior. However, the direct evidence for FM spin fluctuations from polarized inelastic neutron scattering (INS) experiments is still absent [9]. This prevents further understanding of the correlation between AFM and FM spin fluctuations in FeSCs.

The recent progress on bulk superconductivity in iron germanide compound YFe$_2$Ge$_2$ with $T_c$ below 1.8 K shed light on the above issue [10,11]. YFe$_2$Ge$_2$ has the same crystal structure as the 122-structure family of FeSCs and is isoelectronic to the maximally hole-doped KFe$_2$As$_2$, as shown in the inset of Fig. 1. Due to the existence of Ge-Ge bonds, the Fermi surface geometry of YFe$_2$Ge$_2$ resembles that of KFe$_2$As$_2$ under high pressure [12,13], which has a similar collapsed tetragonal phase (CTP) as the existence of As-As bonds. All these facts suggest that YFe$_2$Ge$_2$ is a good reference compound of KFe$_2$As$_2$ to investigate the correlation between AFM and FM spin fluctuations. Theoretically, the standard density functional theory (DFT) calculation predicted that...
the magnetic ground state of YFe₂Ge₂ is an A-type order with a dominated in-plane ferromagnetic correlation \[13\]. However, experimentally, there is no evidence for such magnetic order in YFe₂Ge₂ \[10,11,14\]. Only a large fluctuating magnetic moment on the Fe atom was revealed by the x-ray spectroscopy experiment \[15\]. The recent NMR experiment on YFe₂Ge₂−xSi₇ polycrystalline samples also supported the existence of FM spin fluctuations \[16\]. These findings support that YFe₂Ge₂ is close to a magnetic quantum critical point (QCP) with a predominant in-plane ferromagnetic correlation \[13\]. Very recently, unambiguous evidence for the coexistence of stripe-type and A-type (in-plane FM correlation) spin fluctuations from INS experiments has been successfully found in YFe₂Ge₂ single crystals \[9\]. The A-type spin fluctuations were enhanced and became predominant at low temperature. Here, in order to further understand the magnetic ground state and magnetic QCP in YFe₂Ge₂, we conduct an \(^{89}\)Y NMR study on the single crystals, which are from the same sample batch for the recent INS experiment \[9\]. An incoherent-to-coherent crossover due to Hund’s coupling induced electronic correlation is unambiguously revealed, which has already been observed in KFe₂As₂ \[17–19\]. Interestingly, our results also indicate that the low-temperature enhancement of A-type spin fluctuations observed by the INS experiment is tightly bounded to the low-temperature coherent state. Therefore, we conclude that, below the crossover temperature, the YFe₂Ge₂ system is approaching an itinerant magnetic QCP. Our results shed new light on understanding the correlation between AFM and FM spin fluctuations in FeSCs.

II. METHOD

High-quality YFe₂Ge₂ single crystals were synthesized by the tin-flux method \[9\]. The present NMR measurement on \(^{89}\)Y nuclei is conducted from 2 to 300 K. The external magnetic field of 16 T is applied parallel to either the c axis or the ab plane. As shown in Fig. 1, the linewidth shows a weak temperature dependence and is \(~10\) kHz at low temperature. Compared with previous NMR results on polycrystalline samples \[16\], this narrow linewidth indicates that the single crystal used in the present NMR study is of very high quality.

III. RESULTS

First, clear experimental evidence for electronic crossover behavior is observed in YFe₂Ge₂ by both bulk magnetic susceptibility and Knight shift measurement. In general, the temperature-dependent Knight shift can be expressed as \(K(T) = K_{orb} + A_{hf} \chi_{bulk}(T)\), where \(K_{orb}\) is a \(T\)-independent orbital shift, \(A_{hf}\) is the hyperfine coupling tensor between nuclear spins and electron spins, and \(\chi_{bulk}\) is the uniform spin susceptibility. When there is only one spin degree of freedom, the Knight shift \(K(T)\) can be scaled with the bulk susceptibility \(\chi_{bulk}(T)\) and both of them show a similar temperature dependence. As shown in Fig. 2(a), the Knight shift of \(^{89}\)Y exhibits a similar electronic crossover behavior as that in AFe₂As₂ \((A = K, Rb, and Cs)\) family \[17\]. Above \(T^*\) \(~75 \pm 15\) K, the Knight shift \(K(T)\) increases with decreasing temperature, which is consistent with the bulk magnetic susceptibility \(\chi_{bulk}\) in Fig. 2(b). Such temperature-dependent behavior in both \(K(T)\) and \(\chi_{bulk}(T)\) served as evidence of local moments \[17,18\]. Below \(T^*\), the Knight shift gradually decreases with further lowering temperature and then becomes saturated below 16 K. Except for a Curie-tail behavior at low temperature, the bulk magnetic susceptibility
The similar electronic crossover and Knight shift anomaly have already been observed in AFe$_2$As$_2$ ($A = \text{K, Rb, and Cs}$) family [17,18], which are ascribed to an incoherent-to-coherent crossover due to the Hund’s coupling [18]. In this picture, the Hund’s coupling induced orbital-selective electronic correlation can naturally explain the multiple spin degrees of freedom suggested by the Knight shift anomaly [17,19]. The great similarity of the electronic crossover behavior between YFe$_2$Ge$_2$ and AFe$_2$As$_2$ ($A = \text{K, Rb, and Cs}$) strongly suggests that a similar physical scenario is also suitable for YFe$_2$Ge$_2$. This also qualifies the YFe$_2$Ge$_2$ as a reference system to understand AFe$_2$As$_2$ ($A = \text{K, Rb, and Cs}$) family. In addition, the similar temperature-dependent behavior of $K(T)$ was also observed in the early NMR measurement on a polycrystalline sample [16].

On the other hand, previous studies indicated that YFe$_2$Ge$_2$ is close to a magnetic QCP with a predominate in-plane ferromagnetic correlation [13]. In order to further study the critical spin fluctuations due to magnetic QCP in YFe$_2$Ge$_2$, we measured the temperature-dependent spin-lattice relaxation rate $1/T_1$ of $^{89}$Y nuclei. In general, the $1/T_1$ can be expressed in terms of the imaginary part of the dynamic spin susceptibility, $\text{Im}[\chi(\omega_N, \mathbf{q})]$, as

$$
\frac{1}{T_1} = \lim_{\omega_N \to 0} \frac{1}{2} k_B T \sum_{\omega, \mathbf{q}} F_0(q) \frac{\text{Im}[\chi^{\alpha\alpha}(\omega_N, \mathbf{q})]}{h \omega_N},
$$

where the sum is over the wave vector $\mathbf{q}$ within the first Brillouin zone. $\text{Im}[\chi^{\alpha\alpha}(\omega_N, \mathbf{q})]$ is the imaginary part of the dynamic spin susceptibility of electrons at the wave vector $\mathbf{q}$ and with the Larmor frequency $\omega_N$. $F_0(q)$ is the $\mathbf{q}$-dependent form factor, which is a function of the hyperfine coupling tensor $\mathbf{A}(\mathbf{q})$ [22,27]. As shown in Fig. 4(a), the temperature-dependent $1/T_1 T$ slightly increases with decreasing temperature above $T^*$ and then shows a clear decrease below $T^*$. The whole temperature dependence of $1/T_1 T$ is quite consistent with the temperature dependence of the Knight shift. To simplify the discussion, the spin dynamic susceptibility can be understood as $\chi^{\alpha\alpha}(\omega, \mathbf{q}) = \chi^{\alpha\alpha}_{\text{FL}}(\omega, \mathbf{q}) + \chi^{\alpha\alpha}_{\text{AF}}(\omega, \mathbf{q})$, where $\chi^{\alpha\alpha}_{\text{FL}}$ stands for a weakly $\mathbf{q}$-dependent contribution as conventional Fermi liquid and $\chi^{\alpha\alpha}_{\text{AF}}$ stands for a strongly $\mathbf{q}$-dependent contribution from the critical spin fluctuations at some $\mathbf{q}$-vector [28,29]. When only considering the Fermi-liquid-like contribution, the $1/T_1 T$ would roughly follow a similar temperature-dependent behavior as the Knight shift due to the well-known Korringa relation [28]. However, as the critical spin fluctuations at a certain $\mathbf{q}$-vector come in, the $1/T_1 T$ would be enhanced and break the Korringa relation. When the contribution from the critical spin fluctuations dominates, the temperature-dependent behavior of the $1/T_1 T$ would be different from the Knight shift. Therefore, our above results on $1/T_1 T$ suggests the absence of a contribution from critical spin fluctuations. This seems to be inconsistent with the proposed magnetic QCP scenario [13]. A possible explanation for this discrepancy is to consider the filtering effect of the form factor $F(q) = 0$. Through a careful analysis of the form factor at $^{89}$Y sites [21], we found that the hyperfine field due to the A-type spin fluctuations with $q = (0, 0, 1.5)$, which are proved as the predominant spin fluctuations by a recent INS experiment [9], is completely canceled with $F(q) = 0$.
as shown in Fig. 4(d). Therefore, the absence of contribution from A-type spin fluctuations in $1/T_1T$ can be ascribed to such a filtering effect. A similar filtering effect of spin fluctuations has also been observed in cuprates, such as $^{89}$Y and $^{17}$O NMR in YBCO [30,31]. By further comparing to the INS results (as shown in Fig. 4), we found that the remarkable enhancement of A-type spin fluctuations perfectly coincides with the reduction of $1/T_1T$ below $T^*$. It means that the electronic crossover around $T^*$ drives the system approaching a magnetic QCP with a predominant A-type spin fluctuation. In addition, as suggested by previous INS experiments, besides the predominant A-type spin fluctuations, there is also a minor stripe-type spin fluctuation with $q = (0.5, 0, 0.5)$ in YFe$_2$Ge$_2$. As shown in Fig. 4(c), there is no filtering effect on the stripe-type spin fluctuations. So the minor stripe-type spin fluctuations should contribute to $1/T_1T$. By analyzing the anisotropy of $1/T_1T$, we have successfully identified the expected stripe-type spin fluctuations (see the details in the Supplemental Material [21]).

**IV. DISCUSSION**

Next, we would like to compare the temperature dependence of $1/T_1T^*$ between YFe$_2$Ge$_2$ and CsFe$_2$As$_2$. The previous studies indicated that the AFe$_2$As$_2$ ($A = \text{K, Rb, and Cs}$) family also approaches a magnetic QCP [32,33]. Previous INS experiments on KFe$_2$As$_2$ found that the predominant AFM spin fluctuations in this family are located at $q = [\pi (1 \pm \delta), 0]$ with $\delta = 0.16$ [34], which will not suffer the filtering effect at the interlayer Cs sites. As shown in the inset of Fig. 4(a), the temperature-dependent $1/T_1T$ at $^{133}$Cs sites shows that a remarkable enhancement of $1/T_1T$ emerges just below the incoherent-to-coherent temperature with $T^* \sim 75 \text{K}$ [17,19]. This result indicates that the remarkable enhancement of spin fluctuations in CsFe$_2$As$_2$ is also driven by the electronic crossover around $T^*$ as that in YFe$_2$Ge$_2$. Both of these facts indicate that the enhanced spin fluctuations below $T^*$ are actually related to an emergent coherent state.

In this sense, the magnetic QCP in these systems should exhibit an itinerant nature. The previous DFT calculations have successfully predicted the critical spin fluctuations in both YFe$_2$Ge$_2$ and the AFe$_2$As$_2$ family from an itinerant picture [13,34,35]. This is also consistent with our present conclusions. Considering the Hund’s coupling induced electronic correlation in these systems, the itinerant picture is not necessary to be correct. A local spin model has also been proposed for understanding the magnetic QCP in AFe$_2$As$_2$ ($A = \text{K, Rb, and Cs}$) [32]. So why does the itinerant picture work so well in these systems? The key point is the Hund’s coupling induced incoherent-to-coherent crossover, which has

![Image](Image)

**FIG. 4.** (a) $1/T_1T$ versus temperature with an external field of 16 T parallel to the $ab$ plane and $c$ axis, respectively. The inset shows the $1/T_1T$ of $^{133}$Cs in CsFe$_2$As$_2$. (b) Temperature dependence of the A-type spin fluctuations with $\chi'(E = 7 \text{meV}, Q = (0.5, 0, 0.5))$. The data are taken from the previous INS measurement [9]. (c), (d) Schematic illustration of the transferred hyperfine fields at $^{89}$Y sites. The sources of the hyperfine field come from stripe-type spin fluctuations $(0.5,0,0.5)$ as shown in (c). The sources of the hyperfine field come from A-type spin fluctuations $(0,0,1.5)$ as shown in (d). Violet spheres represent the Fe atoms and small blue spheres represent $^{89}$Y nuclei. The red bold arrows represent the magnetic moment on Fe sites. The green and red thin arrows represent the hyperfine fields from upper and lower Fe-Ge planes, respectively.
a very similar role as the Kondo crossover in heavy fermion systems [36]. In heavy fermion systems, the nature of magnetic QCP (local or itinerant) also strongly depends on the Kondo crossover [37]. When the magnetic QCP is located inside the Kondo crossover, it is always itinerant in nature, the same as YFe$_2$Ge$_2$ and AF$_2$As$_2$ ($\Lambda = K, Rb$, and Cs). In addition, a similar correlation between FM spin fluctuations and electronic crossover was also observed in Sr$_2$RuO$_4$, in which the entire electronic system also develops into a coherent state accompanied by the growth of low-energy FM spin fluctuations in the RuO$_2$ plane [38]. The Hund’s coupling induced orbital-selective electronic correlation also plays a key role in this case, suggesting a universal picture among all these materials [39].

On the other hand, after the confirmation of the A-type spin fluctuations with in-plane FM correlation in YFe$_2$Ge$_2$, a natural question is how to understand the interplay between the in-plane FM spin fluctuations and superconductivity in YFe$_2$Ge$_2$. The previous angle-resolved photoemission spectroscopy result suggests that the electron-phonon coupling should be taken into account for the pairing mechanism in YFe$_2$Ge$_2$ [40]. If the superconductivity is really induced by spin fluctuations in an opposite manner. In this case, the coexistence of AFM and FM spin fluctuations may also lead to a low $T_c$. In addition, the predominant FM spin fluctuations below $T^*$ strongly suggest the pairing mechanism in YFe$_2$Ge$_2$ might favor a spin-triplet pairing, which is consistent with previous electronic structure calculations [13]. This still needs more experiments to confirm, such as a Knight shift measurement below $T_c$. Considering the similar Fermi surface geometry between YFe$_2$Ge$_2$ and the CTP of AF$_2$As$_2$ ($\Lambda = K, Rb$, and Cs), the enhanced FM spin fluctuations may also exist in the CTP of AF$_2$As$_2$. If this is true, then the nonmonotonic behavior of $T_c$ in AF$_2$As$_2$ under pressure can be related to the competition between AFM and FM spin fluctuations [44–46]. A possible spin-triplet pairing is also expected in the CTP of AF$_2$As$_2$. In conclusion, the present work indicates that YFe$_2$Ge$_2$ provides a good platform to study the relation between spin fluctuations and superconducting pairing in FeSCs. Moreover, a potential spin-triplet superconductivity may exist in both YFe$_2$Ge$_2$ and AF$_2$As$_2$ under high pressure.

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