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Persistence of slow fluctuations in the overdoped regime of $\text{Ba}(\text{Fe}_{1-x}\text{Rh}_x)_2\text{As}_2$ superconductors

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We present nuclear magnetic resonance evidence that very slow (≤ 1 MHz) spin fluctuations persist into the overdoped regime of $\text{Ba}(\text{Fe}_{1-x}\text{Rh}_x)_2\text{As}_2$ superconductors. Measurements of the ^{75}As spin echo decay rate, obtained both with Hahn Echo and Carr Purcell Meiboom Gill pulse sequences, show that the slowing down of spin fluctuations can be described by short-range diffusive dynamics, likely involving domain walls motions separating $(\pi/a, 0)$ from $(0, \pi/a)$ correlated regions. This slowing down of the fluctuations is weakly sensitive to the external magnetic field and, although fading away with doping, it extends deeply into the overdoped regime.

INTRODUCTION

The understanding of the electronic properties of iron-based superconductors has significantly progressed over the last years. Superconductivity arises on the verge of an ordered magnetic phase with wavevector $(\pi/a, 0)$ (or $(0, \pi/a)$), characterized by an orthorhombic distortion and by a population imbalance between d_{xz} and d_{yz} Fe orbitals [1]. Most of the debate is now focused on determining how the lattice, spin and orbital degrees-of-freedom intertwine [2]. Nonetheless, a detailed comprehension of the spin dynamics, that are widely thought to play a central role, is still lacking. In particular, nuclear magnetic resonance (NMR) experiments have suggested that the sharp magnetic transition at low electron doping evolves into a cluster spin-glass behavior near the optimal doping level for superconductivity [3]. More recent NMR and neutron scattering studies [4, 5] gave further support for the presence of a cluster spin-glass phase coexisting with superconductivity at low electron doping. Remarkably, even when long-range magnetic order and cluster spin-glass phases vanish, enhanced low-frequency fluctuations (MHz range) persist in the normal phase of different families of iron-based superconductors [3, 6–10]. The origin of these slow dynamics still remains unsettled.

In general, spin dynamics may become glassy (i.e. slow and inhomogeneous) under the influence of quenched disorder or in the case of competing interactions [11]. In iron-pnictides, slow fluctuations have been argued to arise from the motion of domain walls [12–14] that separate $(\pi/a, 0)$ and $(0, \pi/a)$ correlated spin fluctuations, a situation analogous to that observed in frustrated vanadates [15]. The slowing down of domain wall fluctuations may be related to pinning driven by quenched disorder [7] or might be intrinsically due to underlying geometric frustration and long-range Coulomb repulsion [16]. A recent theory also argues that phase separation could drive

a glass-like freezing [17], but this implicitly requires the onset of superconductivity whereas low frequency fluctuations are observed to develop already in the normal state.

While the amplitude of slow dynamics must increase on approaching the spin density wave (SDW) transition occurring at low electron doping, it is not yet clear what happens on moving towards the overdoped superconducting regime. In this article, we show from NMR echo decay measurements that very slow spin fluctuations actually persist at least up to 11% doping in the overdoped regime of $\text{Ba}(\text{Fe}_{1-x}\text{Rh}_x)_2\text{As}_2$. Rh doping induces an increase of electron concentration in the conduction bands of BaFe_2As_2 (Ba122) very much akin to Co doping. In fact, Rh and Co-doped Ba122 display practically identical phase diagrams [18, 19].

By combining different spin-echo techniques, we demonstrate that the low-temperature increase in the transverse relaxation rate $1/T_2$ originates from an activated slowing down of the fluctuations, rather than from an increase of their amplitude. Moreover, it is shown that the activated correlation time describing $1/T_2$, with an energy barrier decreasing with Rh doping, accounts also for the behavior of the spin-lattice relaxation rate $1/T_1$. We finally evidence that this type of fluctuations extends at least up to about 11% of doping, differently from earlier results based on $1/T_1$ analysis [3], suggesting that the vanishing of the superconducting phase in $\text{Ba}(\text{Fe}_{1-x}\text{Rh}_x)_2\text{As}_2$ is followed by the concomitant fading of these low-frequency excitations.

MATERIALS AND METHODS

^{75}As NMR experiments were performed on $\text{Ba}(\text{Fe}_{1-x}\text{Rh}_x)_2\text{As}_2$ single crystals [20] with Rh content and superconducting transition temperature (T_c) of

$x = 4.1\%$ ($T_c = 13.6$ K), $x = 6.8\%$ ($T_c = 22.4$ K), $x = 9.4\%$ ($T_c = 15.1$ K) and $x = 10.7\%$ ($T_c = 12.25$ K), respectively. T_c was determined by superconducting quantum interference device (SQUID) magnetometry prior to the NMR experiment and also checked *in situ*, via the observation of the detuning of the NMR tank circuit. The magnetic field \mathbf{H} was applied along the crystallographic c axis, unless otherwise stated.

The echo-decay time was first measured by the standard Hahn echo sequence: $\pi/2 - \tau - \pi$ [21]. Since the spin-lattice relaxation time T_1 and the raw Hahn echo decay time have both values in the 1-100 ms range, one can expect a sizable contribution of T_1 processes to the echo decay (Redfield term [22, 23]), as confirmed by the linear dependence of the raw echo decay rate $1/T_2^t$ on $1/T_1$ at high temperature (Fig. 1 (a)). In fact, the Hahn echo decay $M_t(2\tau)$ can be written [24]:

$$M_t(2\tau) = M(2\tau) \exp\left(-\frac{2\tau}{T_{1R}}\right) \quad (1)$$

evidencing that the relaxation involves both spin-lattice relaxation processes, via the T_{1R} term, and a T_1 independent $M(2\tau)$ term. In case of an anisotropic spin-lattice relaxation rate, Walstedt and coworkers [24] obtained a general result for the central ($\frac{1}{2} \rightarrow -\frac{1}{2}$) transition of half integer spin, which for $I=3/2$ is:

$$\frac{1}{T_{1R}^{\parallel}} = \frac{3}{T_1^{\parallel}} + \frac{1}{T_1^{\perp}} \quad (2)$$

where the symbols \parallel and \perp refer to the magnetic field orientation with respect to the crystallographic c axis. Once the raw echo decay data have been corrected for the spin-lattice relaxation term, the Hahn echo decay contribution $M(2\tau)$ was analyzed. The Hahn echo decay was found to deviate from a single exponential (Fig. 2(a)) and could be fit in general to a stretched exponential, $M(2\tau) = M_0 \exp(-(2\tau/T_2)^\beta)$, where the stretched exponent showed a marked temperature dependence (Fig. 2(b)).

The echo decay time was also measured with the Carr-Purcell-Meiboom-Gill (CPMG) sequence [25], in which the $\pi/2$ pulse is followed by a comb of π pulses, separated by a time τ_{cpmg} , ranging between 80 μs and 500 μs (Fig. 2(a)). The echo amplitude decays exponentially with time and the decay rate is found to increase linearly with τ_{cpmg} (Fig. 1(b)). Hence, $1/T_{2cpmg}$ can be conveniently defined by taking the value extrapolated for $\tau_{cpmg} \rightarrow 0$.

In the iron-based superconductors, few works have attempted to measure the spin-echo decay time also with a CPMG sequence [10, 26]. However, a comparative study between Hahn and CPMG sequences is beneficial in revealing the presence of slow spin dynamics. At high temperature ($T \geq 50$ K), the spin-echo decay measured by both methods is temperature independent, with

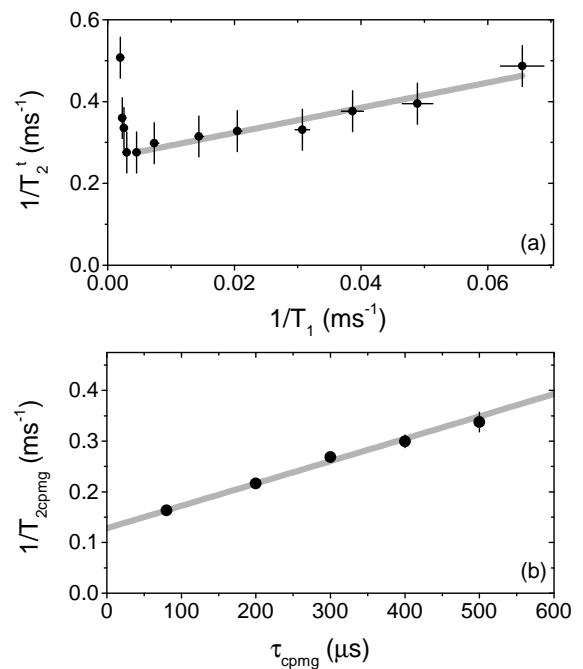


FIG. 1. (a) The raw Hahn echo decay rate (T_2^t) versus the spin-lattice relaxation rate in the normal phase of the $x = 9.4\%$ sample at $H = 6.4$ T. T_2^t is defined as the time at which the normalized echo amplitude decays to $1/e$. The temperature is an implicit parameter. The grey line is the linear fit to the data above 20 K. (b) $1/T_{2cpmg}$ plotted as a function of τ_{cpmg} , for the compound $x = 6.8\%$, measured at 6.4 T, and 70 K. The grey line is a fit to extrapolate the intrinsic T_{2cpmg} value.

$1/T_{2cpmg} < 1/T_2$ (Fig. 2(c)), and smaller than the value $1/T_2^{dip} = 1.4 \text{ ms}^{-1}$ expected from the dipolar interaction between As nuclei [6]. The origin of this discrepancy will be discussed subsequently. Since the π pulses of the CPMG sequence were not phase alternated, T_{2cpmg} could be affected by spin-locking effects [27–29]. This could explain the difference between T_2 and T_{2cpmg} at high temperature where both times are T independent, but it does not affect our conclusions concerning the different T-dependence observed at low temperature.

Remarkably, $1/T_2$, measured by the Hahn echo sequence, shows a pronounced enhancement starting above T_c , on cooling (Fig. 2(c)). This increase, observed at all magnetic fields (Fig. 3), is not detected by the CPMG echo sequence (Fig. 2(c)). This dichotomy is observed in all the studied samples, thus corroborating and complementing the initial findings of Ref. [6].

Finally, we point out that the spin-lattice and transverse relaxation times are not homogeneous across the NMR line. The results in Fig. 4 show two representative plots for the overdoped sample with $x = 10.7\%$, at 17 T. The T_2 and T_1 variation is $\sim 65\%$, across the whole spectrum. This spectral distribution of relaxation times suggests that not all spins have the same spin temperature. The values of $1/T_1$ and $1/T_2$ reported in this

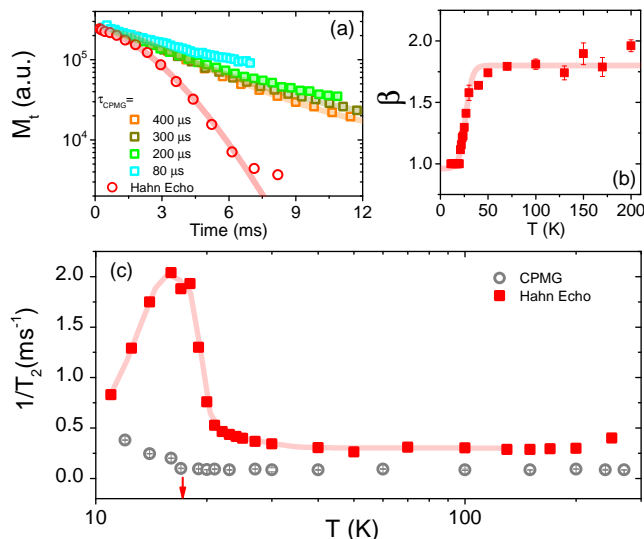


FIG. 2. **(a)** Hahn (red circles) and CPMG echo decays, measured on the $x=6.8\%$ compound, at $T = 70$ K and $H = 6.4$ T. The delay τ_{cpmg} between CPMG echoes is indicated. The comparison between the two pulse sequences evidences the deviation from single exponential in the Hahn echo decay. The solid lines are the fit to the equations discussed above. **(b)** The stretched exponent is reported as a function of the temperature, measured for the same sample, at 11 T. The shadowed line is a guide to the eye. **(c)** Hahn (red squares) and CPMG (grey circles) echo decay rates, as a function of the temperature, measured in a magnetic field of 11 T. The raw data have been corrected for the Redfield term. The arrow marks T_c . The red line is a guide for the eye.

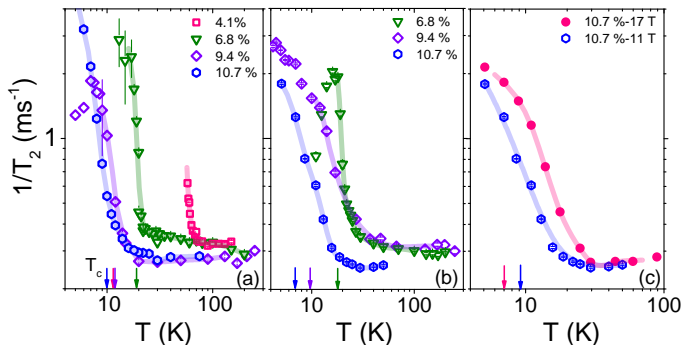


FIG. 3. Hahn Echo decay rates as a function of the temperature, for $\text{Ba}(\text{Fe}_{1-x}\text{Rh}_x)_2\text{As}_2$ for different magnetic fields: **(a)** $H=6.4$ T, **(b)** $H=11$ T and **(c)** for $x=10.7\%$ at 11 and 17 T. The arrows mark T_c . The shadowed lines are guides to the eye.

manuscript were recorded irradiating the central part of the spectrum. We notice that the same spatial magnetic inhomogeneity was also observed in the T_1 measurements of the Co-doped compounds [7, 30].

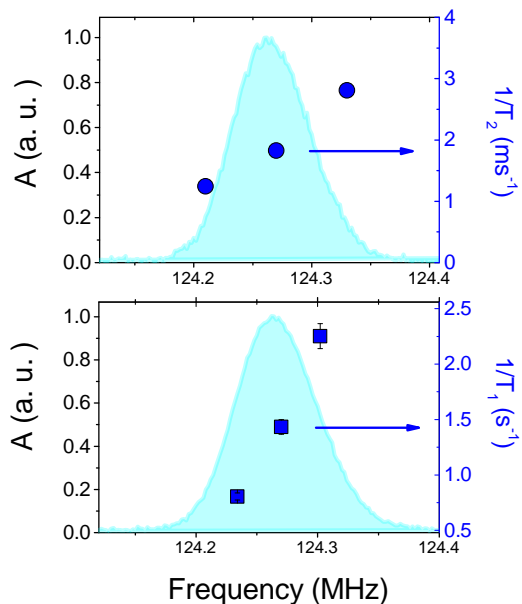


FIG. 4. **(top)** $1/T_2$ (right axis) as a function of the frequency, measured at 17 T and 150 K, for the $x = 10.7\%$ sample. **(bottom)** $1/T_1$ (right axis) as a function of the frequency, measured at 17 T and 7 K, for the same sample. The left axes represent the spectral intensity, in arbitrary units.

DISCUSSION

Let us first consider the difference between the Hahn and CPMG sequences. While the former is very effective in refocusing the dephasing of the in-plane nuclear magnetization coming from *static* field inhomogeneities, the amplitude of the Hahn echo is decreased by dynamics with a fluctuation time scale of the order of the separation between the $\pi/2$ and π pulses. In case of diffusive-like dynamics in a field gradient $\nabla\mathbf{B}$, described by a diffusion coefficient D , one should weight $M(2\tau)$ by $\sim \exp(-\gamma^2|\nabla\mathbf{B}|^2D(2\tau)^3)$. Carr and Purcell (CP) [25] devised a way to quench the effects of these dynamics by slicing the time τ over which the dynamics would irreversibly quench the echo amplitude with many π pulses, separated by $\tau_{cpmg} \ll \tau$. Accordingly, the dynamics would become effective in reducing the echo amplitude only if its characteristic time scale is of the order of τ_{cpmg} . The original CP sequence was later implemented into the CPMG one, in order to avoid phase error accumulation.

Now, in Rh-doped Ba122 compounds we observed a linear increase of $1/T_{2cpmg}$ with τ_{cpmg} , which is typical of systems, such as platinum nanoparticles [31], where restricted electron spin diffusion in a non-uniform magnetic field occurs [10, 23, 26, 32]. In the $\tau_{cpmg} \rightarrow 0$ limit, $1/T_{2cpmg}$ is no longer affected by the dynamics and only the irreversible decay due to nuclear dipole-dipole interaction between ^{75}As nuclei should be effective. This intrinsic decay time, $T_{2i} \sim 10$ ms, should be compared

with the much shorter one estimated from lattice sums for ^{75}As - ^{75}As dipolar interaction, equal to 0.7 ms. The long experimental value of T_{2i} suggests that not all the As nuclei are contributing to the dipolar field distribution, as in presence of a mechanism quenching the nuclear spin flip-flop mechanism [23]. The suppression of the latter occurs when the inhomogeneous NMR linewidth is much larger than the dipolar coupling between ^{75}As nuclei, as justified below. The quenching of flip-flop mechanisms is further supported by the distribution of relaxation rates observed across the NMR line (Fig. 4), indicating the absence of a common spin-temperature among ^{75}As nuclei and suggesting that the electronic system is highly inhomogeneous.

Unlike $1/T_{2cpmg}$ ($\tau_{cpmg} \rightarrow 0$), the Hahn echo decay rate $1/T_2$ is sensitive to electron spin diffusive dynamics. Given the inhomogeneous nature of the electronic texture [33, 34], an internal magnetic field gradient $\nabla\mathbf{B}$ could originate from a spatial inhomogeneity of the spin susceptibility $\Delta\chi$ or, equivalently, of the local magnetization $\Delta\chi H$ [35, 36]. Hence, $\nabla\mathbf{B} \simeq \Delta\chi\mathbf{H}/2a$, where $2a$ defines a typical domain size [35]. A successful approach to treat the echo relaxation in the case of restricted diffusion was presented by Robertson [32, 37]. Robertson showed that it is possible to describe restricted spin diffusion by an equivalent mechanism of unrestricted diffusion in a periodic field gradient. As mentioned above, we assume that the source of *internal* field inhomogeneity here comes from the distribution of hyperfine fields at ^{75}As nuclei, affecting the NMR linewidth $\Delta\nu$. Thus, we can write the field gradient probed by the nuclei as

$$\nabla\mathbf{B}_{hyp} = \frac{\pi\Delta\nu}{a\gamma} \quad (3)$$

where γ is the nuclear gyromagnetic ratio. From the equation above, by taking a linewidth of 30 kHz [6], and $2a$ equal to few lattice steps [38, 39], the internal field gradient results $\nabla B \sim 10^8$ G/cm. Therefore two As nuclei separated by 0.6 nm experience a Larmor frequency difference of about 4 kHz, which is much larger than the dipolar interaction, estimated from lattice sums to be ~ 200 Hz. This justifies the fact that nuclear spin flip-flop processes are quenched [40].

Assuming that the periodicity of the field gradient is equal to the diffusion length, we can write for the Hahn echo decay rate [32]:

$$\frac{1}{T_2}(T) \simeq \frac{(\pi a \Delta\nu(H, T))^2}{120D(T)} + \frac{1}{T_{2i}} \quad (4)$$

where T_{2i} is the intrinsic relaxation time in absence of dynamics. D is the spin-diffusion coefficient directly related to the characteristic fluctuation time $\tau_D = a^2/D(T)$ [25]. From the raw $\Delta\nu$ [6], it is then straightforward to derive τ_D . The spin-diffusion time can be fitted to an Arrhenius law $\tau_D(T) = \tau_0 e^{U/T}$ (Fig. 5), with $\tau_0 = 1$ -100 ns.

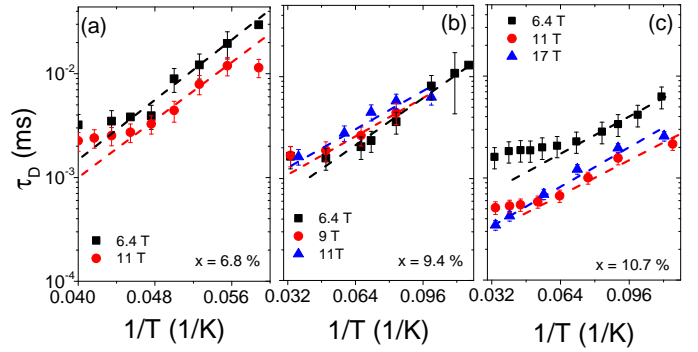


FIG. 5. Diffusion time for different Rh concentrations, at different magnetic field strengths. The dashed lines are linear fit to the Arrhenius law, as described in the text.

We notice that the field dependence of $1/T_2$ reported in Fig. 3(c) cannot be ascribed to the weak field dependence of τ_D (see Fig. 5 (c)) but should mainly be associated with the field dependence of the linewidth. We found that the energy barrier U decreases nearly exponentially with Rh doping (Fig. 6), and, as shown in Fig.5, it is weakly affected by the magnetic field. Moreover we notice that, being $\tau_D = 0.1 - 1 \mu\text{s}$, the condition of applicability of Eq. (4), namely $\tau \gg a^2/D$, is satisfied [32]. We also notice that the field dependence of $1/T_2$ observed in our previous work [6] is here justified by Eq. 4, where the linewidth explicitly enters into the Hahn echo decay time.

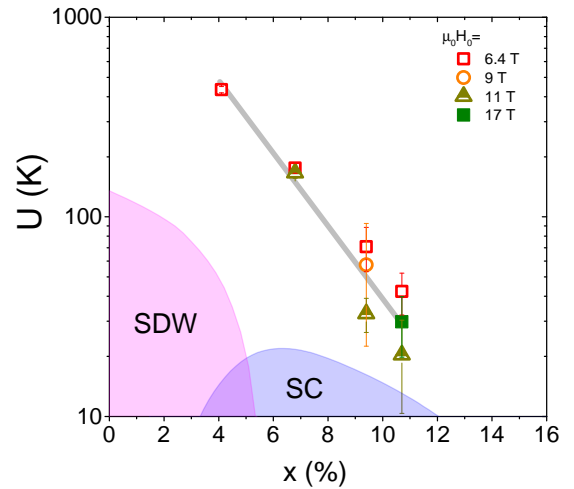


FIG. 6. Activation barrier U as a function of Rh content (x) for different magnetic fields. The energy barrier displays an exponential decrease with doping (grey line). The effect of the magnetic field is weak in almost all samples. The shadowed domes represent the superconducting and Spin Density Wave regions of the phase diagram.

The model of restricted spin diffusion can shed light also on the temperature dependence of the spin-lattice relaxation rate. The latter slightly deviates from linearity

above T_c , evidencing the presence of weak magnetic correlations [41, 42]. Additionally, the optimally doped and weakly overdoped compounds display a hump in the spin-lattice relaxation rate, above T_c [6, 9], when the magnetic field is oriented in-plane ($\mathbf{H} \perp c$). If we assume that the spin diffusion is associated with random fluctuating local fields, which can be described by a correlation function $g(t) = h_0 e^{-t/\tau_D}$, the spectral density of spin fluctuations at the Larmor frequency ω_L [23] leads to:

$$\frac{1}{T_1} = A \frac{(2\pi\Delta\nu)^2}{1 + (\omega_L\tau_D)^2} \tau_D + BT^b \quad (5)$$

The first term corresponds to the so-called Bloembergen-Purcell-Pound (BPP) model [43, 44], with root mean-squared value of the transverse field equal to $2\pi\Delta\nu/\gamma$ and correlation time for the field fluctuation equal to the diffusion time τ_D . The second term in Eq. 5 accounts for the weakly correlated electron spin dynamics and for deviations from the Korringa law. The fit in Fig. 7 is obtained from three parameters, τ_D , B and b , where the latter two can be fixed from the high temperature regime. Despite its simplicity, this model captures the essential features of the experimental results, except around T_c , owing to the opening of the superconducting gap. Furthermore, the fit in Fig. 7 returns an energy barrier of $U \simeq 50$ K which agrees with that derived from the analysis of $1/T_2$ (see Fig. 6 for comparison). Therefore, the hump in $1/T_1$ can be attributed to the very same diffusive-like dynamics that give rise to the enhancement in $1/T_2$.

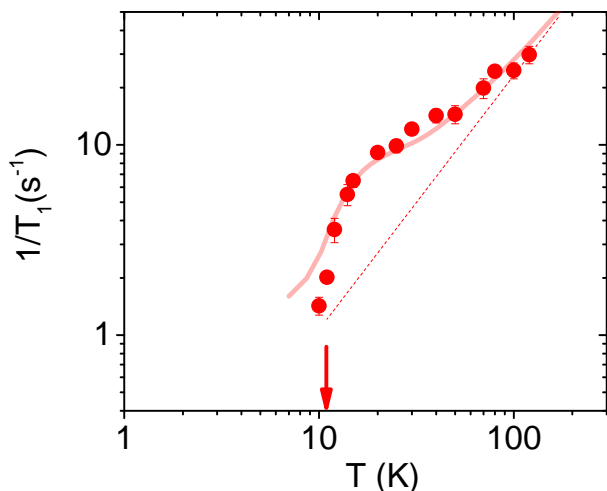


FIG. 7. Spin-lattice relaxation rate as a function of the temperature for the $x = 9.4\%$ sample, measured with the in-plane field ($\mathbf{H} \perp c$). The solid line is the fit according to Eq. 5 in the text. The dashed line represents the spin-lattice relaxation rate behavior for $\mathbf{H} \parallel c$.

Analogous activated behavior with similar U values have been reported in $\text{LaFeAsO}_{1-x}\text{F}_x$ [8, 9]. However, in contrast to $\text{LaFeAsO}_{1-x}\text{F}_x$, here the energy barrier is

significantly doping dependent, with a marked decrease upon increasing the Rh content (Fig. 6). A natural question that arises is whether this energy scale goes to zero at a finite doping level and whether this doping defines a quantum critical point (QCP) [45]. We cannot fully address this question here, but the close values of U for the samples with 9.4% and 10.7% samples do not point towards a QCP associated with these dynamics, which seem rather to persist in the overdoped regime and slowly fade away with superconductivity.

We found that the low-temperature increase of ^{75}As $1/T_2$ in $\text{Ba}(\text{Fe}_{1-x}\text{Rh}_x)_2\text{As}_2$ is not associated with an increase in the amplitude of the spin fluctuations, as it is the case for ^{63}Cu $1/T_2$ in the normal phase of superconducting cuprates [46]. Here, $1/T_2$ increases mostly due to a slowing down of the dynamics to the MHz range. These slow dynamics are also evidenced by a field dependent hump in $1/T_1$ (Fig. 7) [9], while in the cuprates $1/T_1$ is dominated by high frequency correlated spin dynamics yielding only a weak, if any, magnetic field dependence of $1/T_1$ at $T > T_c$ [47, 48]. The only case in cuprates that bears some resemblance to our data is the increase of $1/T_2$ triggered below the onset of charge order [49–51]. However, no evidence of charge order has been found in pnictides. Moreover, the experimental evidence that in $\text{Ba}(\text{Fe}_{1-x}\text{Rh}_x)_2\text{As}_2$ these low frequency fluctuations extend from the underdoped to the overdoped regime rather suggests that the normal phase of iron-based superconductors is characterized by unconventional excitations which are absent in the cuprates.

Even if electron-doped iron based superconductors are generally considered as itinerant systems with moderate electron correlations, the $J_1 - J_2$ model has been shown to effectively provide an insightful approach to describe some of their magnetic properties. In particular, in the prototypes of $J_1 - J_2$ model, it has been observed that, similarly to what we found here, slow dynamics develop for $T < J_1 + J_2$, at frequencies several orders of magnitude below $k_B(J_1 + J_2)/\hbar$ [15]. This has been ascribed to activated fluctuations of domain walls separating regions with $(\pi/a, 0)$ and $(0, \pi/a)$ correlations. The correspondent energy barrier agrees with the theoretical prediction by Chandra, Coleman and Larkin [52]. More recently, Mazin and Johannes [12] have suggested that such low frequency domain wall excitations should be present also in the iron-based superconductors. Therefore, it is likely that the very slow fluctuations seen here are related to the dynamics of domain walls separating nematic domains with perpendicular magnetic wave-vectors. Within that framework, the energy barrier should scale with the square of the in-plane electron spin correlation length [52] and the decrease of U would indicate a decrease of electron correlations with electron doping [53].

While evidence for spin nematic and orbital nematic fluctuations, even well above the ordering temperature,

have been reported in the underdoped regime of iron-based superconductors [38, 54–56], no clear evidence for the persistence of slow fluctuations driven by nematicity has been presented for the overdoped iron-based superconductors. It is interesting to notice that the vanishing of the spin fluctuations probed by $1/T_2$ is accompanied by a decrease in the amplitude of charge fluctuations of nematic character probed by inelastic Raman scattering [57], as well as by a decrease of the orbital anisotropy [58]. The persistence of nematic fluctuations in the overdoped regime [57] appears consistent with our finding of slow fluctuations remaining well above optimal doping.

CONCLUSIONS

By measuring the spin echo decay rate with different pulse sequences, we have evidenced the presence of low-frequency fluctuations developing in the normal phase of $\text{Ba}(\text{Fe}_{1-x}\text{Rh}_x)_2\text{As}_2$ iron-based superconductors. The comparison between $1/T_{2cpmg}$ and $1/T_2$ has suggested the presence of restricted spin diffusive dynamics. Within this framework, the behavior of $1/T_2$ and of $1/T_1$ can be analyzed consistently and the fluctuations can be described by an activated correlation time with an energy barrier exponentially decreasing with Rh-doping. Our results point out that very slow spin dynamics persist into the overdoped regime and could be tentatively associated with domain walls fluctuations. These dynamics, which are an indirect consequence of the presence of nematic correlations, are likely to be observed in all electron doped iron-based superconductors.

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