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Abstract
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Evidence of unconventional low-frequency dynamics in the normal phase of Ba(Fe\textsubscript{1−x}Rh\textsubscript{x})\textsubscript{2}As\textsubscript{2} iron-based superconductors

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This work presents \textsuperscript{75}As NMR spin-echo decay rate (1/T\textsubscript{2}) measurements in Ba(Fe\textsubscript{1−x}Rh\textsubscript{x})\textsubscript{2}As\textsubscript{2} superconductors, for 0.041 ≤ x ≤ 0.094. It is shown that 1/T\textsubscript{2} increases upon cooling, in the normal phase, suggesting the onset of an unconventional very low-frequency activated dynamic. The correlation times of the fluctuations and their energy barriers are derived. The motion is favored at large Rh content, while it is hindered by the application of a magnetic field perpendicular to the FeAs layers. The same dynamic is observed in the spin-lattice relaxation rate, in a quantitatively consistent manner. These results are discussed in the light of nematic fluctuations involving domain wall motion. The analogies with the behavior observed in the cuprates are also outlined.

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The study of the excitations in the normal phase of superconductors (SC) is of major importance to unravel the mechanisms driving the Cooper pair formation. Both in the cuprates and in the iron pnictides the presence of competing interactions gives rise to complex phase diagrams and to quasidegenerate ground states, which can induce unconventional dynamics at low energies. NMR has played a key role in the study of low-frequency (LF) excitations in the normal state both of high-T\textsubscript{c} SC and, more recently, of the iron-based SC. Most of the NMR investigations carried out so far in these materials have concentrated on the dynamical features emerging from the spin-lattice relaxation rate (1/T\textsubscript{1}) measurements,\textsuperscript{1–7} while less attention has been paid to the study of the spin-echo decay rate 1/T\textsubscript{2}, which is quite a useful tool to probe very LF excitations.\textsuperscript{8–10} In the cuprates, one of the most significant achievements was the derivation of the staggered static spin susceptibility from \textsuperscript{63}Cu(2) Gaussian echo decay rate 1/T\textsubscript{2G}.\textsuperscript{11,12} However, other nuclei show a different behavior of 1/T\textsubscript{2}: \textsuperscript{89}Y NMR decay rate, in the SC phase of YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7}, presents an exponential term,\textsuperscript{13,14} which reveals a peak in 1/T\textsubscript{2}, that was first ascribed to vortex dynamics.\textsuperscript{15} Remarkably, \textsuperscript{17}O NMR revealed a second peak that, however, was also seen in \textsuperscript{63}Cu nuclear quadrupole resonance experiments in YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7−x}, where no magnetic field was applied, thus questioning the former explanation and suggesting other mechanisms involving charge fluctuations.\textsuperscript{14–16}

Similar trends of 1/T\textsubscript{2} have also been reported in the recently discovered iron-based SC. In the optimally doped Ba(Fe\textsubscript{1−x}Co\textsubscript{x})\textsubscript{2}As\textsubscript{2} (Ref. 9) and in Ba(Fe\textsubscript{1−x}Rh\textsubscript{x})\textsubscript{2}As\textsubscript{2} (BaFeRh\textsubscript{122} hereafter),\textsuperscript{9} a peak in 1/T\textsubscript{2} was detected below T\textsubscript{c} and again associated with the vortex dynamics. On the other hand, the behavior of \textsuperscript{75}As NMR T\textsubscript{2} in the normal phase of BaFeRh\textsubscript{122} SC is not completely understood\textsuperscript{8,9} and its magnitude is far from any theoretical expectation.

In the following, a systematic study of \textsuperscript{75}As NMR spin-echo decay in BaFeRh\textsubscript{122} iron-based SC, over a broad range of Rh doping, is presented. The echo shows a high temperature (T) dominant Gaussian decay which becomes exponential at low T. The exponential decay rate increases upon cooling already in the normal phase, suggesting the onset of an unconventional very LF activated dynamics, whose characteristic correlation times are derived together with the corresponding energy barriers. This dynamic persists across the whole phase diagram up to the overdoped compounds, but it is less pronounced if the magnetic field is applied perpendicularly to the FeAs layers. It is also shown that the same dynamics affect 1/T\textsubscript{1}. These LF fluctuations are discussed in terms of domain wall motion, possibly involving nematic fluctuations.

NMR measurements have been performed on three BaFeRh\textsubscript{122} single crystals: an underdoped sample, with x = 4.1% (T\textsubscript{c} = 13.6 K), a nearly optimally doped sample, with x = 6.8% (T\textsubscript{c} = 22.4 K), and an overdoped sample, with x = 9.4% (T\textsubscript{c} = 15.1 K). The samples were grown as outlined in Ref. 17. The critical temperature T\textsubscript{c} was determined via superconducting quantum interference device magnetometry and it is in agreement with Ref. 17. \textsuperscript{75}As NMR experiments were performed at 6.4, 9, and 11 T, for H\textsubscript{0} parallel and perpendicular to the c axis. The spin-echo decay time was estimated by fitting the decay of the transverse nuclear magnetization M\textsubscript{t}, measured either after a standard Hahn echo (HE) sequence (π/2−τ−π) or by using a Carr-Purcell-Meiboom-Gill (CPMG) sequence (π/2−τ\textsubscript{CP}−π−τ\textsubscript{CP}−π−…). In the latter, the delay τ\textsubscript{CP} was varied in order to extract the intrinsic decay time T\textsubscript{2CPMG} for τ\textsubscript{CP} → 0. The HE decay was first corrected in order to remove the contribution of the spin-lattice relaxation rate 1/T\textsubscript{1}.\textsuperscript{18} Afterwards, the echo decay could be nicely fit by the product of an exponential and a Gaussian decay (inset of Fig. 1):

\[
M(t)/M_0 = \exp(-2\tau/t_2) \exp\left(-\frac{(2\tau)^2}{2T_{2G}^2}\right).
\]

The two components evolve with temperature in such a way that, in the high-T\textsubscript{c} regime, the Gaussian term is significantly larger than the exponential one, and both are weakly temperature dependent. This trend persists down to a temperature T\textsuperscript{*} > T\textsubscript{c}, where the Gaussian contribution becomes negligible, while the exponential rate grows rapidly and becomes the

main contribution to the echo decay (Figs. 1 and 2). The experimental values of $T^*$ are $22 \pm 2$ and $18 \pm 2$ K for $x = 6.8\%$ and $x = 9.4\%$, respectively. Here the low-$T$ exponential component is discussed, since the high-$T$ constant behavior has been discussed by Oh et al.\(^9\) From Fig. 3 one also notices that $1/T_{2\text{CPMG}}$ is significantly reduced with respect to $1/T_{2\text{exp}}$ for $T < T^*$ and shows a less pronounced field and $T$ dependence. The HE and CPMG sequences are most sensitive to fluctuations with a characteristic time scale $\tau_c \sim \tau$ and $\sim \tau_{CP}$, respectively. Hence, the observed difference between $1/T_{2\text{exp}}$ and $1/T_{2\text{exp}}$ suggests that the correlation time of the fluctuations probed by $^{75}$As nuclei increases above the $\mu$s, below $T^*$. By comparing the measurements performed on the three crystals, for different magnetic field orientations (Fig. 2) and magnitudes (Figs. 2 and 3), one can conclude that the enhancement of $1/T_{2\text{exp}}$ has four main features: (i) it starts above $T_c$, (ii) it is favored by the magnetic field, (iii) it is accentuated for in-plane fields, and (iv) it persists across the whole phase diagram, up to the overdoped compound (Fig. 3).

Further insights into the LF dynamics can be derived from the $T$ dependence of the FWHM of the central $^{75}$As NMR line, obtained from the Fourier transform of half of the echo (Fig. 2). The NMR spectrum displays an inhomogeneously broad line shape with a LF tail, and it gets broader upon cooling. Remarkably, at the same temperature $T^*$ where the echo decay becomes exponential, the linewidth starts to decrease, hence suggesting the onset of LF dynamics which can average out the static frequency distribution probed by $^{75}$As nuclei. Finally, at $T_m < T_c$, the line broadens again (Fig. 2), as expected when the solid/glassy vortex phase sets in.\(^8,9\)

Since a LF dynamic is present one should expect an effect also on $1/T_1$, which probes the spectral density at the nuclear Larmor frequency. In the inset of Fig. 3 is shown $1/T_1$, as derived after a saturation recovery pulse sequence, for both magnetic field orientations, for $x = 9.4\%$. Remarkably, a bump in the spin-lattice relaxation rate was
The reader may notice that the barrier is comparable with the one found from the $1/T_1$ fit. Moreover $U$ is $H_0$ independent, in the explored range, while it clearly depends on the electronic concentration, namely, it decreases by increasing the Rh content. This trend indicates that the higher the electron doping the faster the dynamics. Furthermore the fluctuating longitudinal local field $h_{L}$ shows a continuous increase with the applied field and the doping. It is also noticed that, when the field is perpendicular to the $c$ axis, the enhancement is significantly larger.

The LF dynamics evidenced by $1/T_{2\exp}$ and by the bump in $1/T_1$ cannot be due to the standard correlated electron spin fluctuations or SC fluctuations which typically occur at frequencies orders of magnitude larger than the frequency probed here. One should look for very LF fluctuations as the ones occurring close to a spin or charge freezing or taking place among quasidegenerate ground states. In this respect, one should consider that, owing to the geometry of the relevant exchange couplings, the magnetic properties of the iron pnictides have often been described within an effective $J_1$-$J_2$ model on a square lattice. The ground state of that model is characterized by two degenerate columnar antiferromagnetic ground states corresponding to two nematic phases, and the fluctuations between those two states can give rise to very LF dynamics. In fact in vanadates, which can be considered as prototypes of that model, these LF fluctuations have been detected by $\mu$SR above the magnetic ordering, in a temperature range where the electron spins are already correlated. These dynamics can be associated with domain wall motion separating correlated regions of the two nematic states. Once the two different phases set in, the domain walls can be put into motion if the energy barrier $U$ separating these two phases is overcome (Fig. 4). Note that a similar scenario has also been proposed in a recent study of the magnetic state of CaFe$_2$As$_2$.

On the other hand, the observations of the magnetic field effect on $1/T_2$ and $1/T_1$, suggest that when $H_0 \parallel c$ the fluctuations are reduced. Such an effect recalls the field-induced charge order recently reported in underdoped YBa$_2$Cu$_{6+y}$O$_{6+y}$ SC, where the blocking of the charge order occurs only if $H_0$ is perpendicular to the highly conductive CuO$_2$ layers. Moreover in these systems, Wu et al. found a temperature and magnetic dependence of $1/T_2$ similar to the one reported here for the iron-based SC. In the light of these analogies one could speculate that very LF fluctuations associated with domain wall motion, possibly involving charge stripes, can be present both in the cuprates and in the iron-based SC. However, further experiments are required to support such a scenario.

In conclusion, this paper presents a systematic study of the spin-echo decay rate in 122 iron-pnictides over a broad doping range. A LF spin dynamic is observed above $T_c$ and it is responsible for a bump in the spin-lattice relaxation time, as well as for an enhancement of the exponential component of the spin-echo decay rate. Such dynamics are common at all the doping concentrations, and they get faster in the overdoped regime. Moreover they can be associated with domain wall motion, possibly involving nematic fluctuations. The remarkable analogies with the behavior found in the...
cuprates indicate the need for a deeper investigation, also by other techniques.

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