Comment on “Discovery of slow magnetic fluctuations and critical slowing down in the pseudogap phase of YBa$_2$Cu$_3$O$_y$”

Jeff E. Sonier,1,2

1 Department of Physics, Simon Fraser University, Burnaby, British Columbia, Canada V5A 1S6
2 Canadian Institute for Advanced Research, Toronto, Ontario, Canada M5G 1Z8

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A recent zero-field (ZF) and longitudinal-field (LF) muon spin relaxation ($\mu$SR) study of YBa$_2$Cu$_3$O$_y$ [Jian Zhang et al., arXiv:1709.06799] claims to have detected critical slowing down of magnetic fluctuations near the pseudogap temperature $T^*$, and attribute this to the onset of slow fluctuating domains of intra-unit-cell magnetic order. Here it is argued that the relaxation data displayed in this study are misleading due to an improper account of the nuclear dipole contribution and a failure to acknowledge the occurrence of muon diffusion.

In a ZF or LF $\mu$SR experiment, the measured asymmetry spectrum $A(t)$ is proportional to the time evolution of the muon spin polarization $P_z(t)$ along the initial direction of the muon spin (defined to be the $z$ axis). Each positive muon ($\mu^+$) implanted in the sample senses a resultant field

$$B_\mu = B_{\text{dip}} + B_{\text{con}} + B_{\text{LF}},$$

where $B_{\text{dip}}$ originates from magnetic dipole moments inside the sample, $B_{\text{con}}$ is the Fermi contact field in conductors generated by the net spin density of conduction electrons in contact with the $\mu^+$, and $B_{\text{LF}}$ is the external magnetic field that is applied in the $z$ direction in a LF experiment. In general, $B_{\text{dip}}$ consists of contributions from both nuclear and electronic moments. The electronic dipolar field sensed by the $\mu^+$ may be static or fluctuating. Since the correlation times of the nuclear moments are typically much longer than the muon life time, the resultant nuclear dipolar field sensed by the $\mu^+$ is generally considered to be static.

Zhang et al. make the implicit assumption that the nuclear dipole contribution to the ZF-$\mu$SR signals of YBa$_2$Cu$_3$O$_{6.72}$ and YBa$_2$Cu$_3$O$_{6.95}$ is $T$-independent and well described by what is known as a static Gaussian Kubo-Toyabe (KT) relaxation function. Neither of these assumptions are valid. The reasons why can be found in an earlier ZF-$\mu$SR study of YBa$_2$Cu$_3$O$_y$ (Ref. 3). There it was shown that the nuclear dipole contribution to the ZF-$\mu$SR signal is modified (1) below $T \sim 100$ K by the formation of charge-density-wave order (CDW) in the CuO chains, (2) an apparent structural change near 60 K, and (3) above $T \sim 160$ K by muon diffusion.

The EFG at nuclei in an ionic crystal lattice is modified by the presence of the $\mu^+$ and in metallic systems by its screening cloud of conduction electrons. In YBa$_2$Cu$_3$O$_y$ the local EFG is also modified by the onset of static CDW order in the CuO chains. Measurements of the nuclear quadrupole resonance (NQR) linewidth $\delta\nu_Q$ in YBa$_2$Cu$_3$O$_7$ show an abrupt increase in the local charge distribution around the Cu(1) chain sites and Cu(2) plane sites across $T_c$. Moreover, the variation of $\delta\nu_Q$ with $T$ at the Cu(2) site is nonmonotonic, exhibiting a broad hump near 60 K and a dip near 40 K. The in-plane charge modulation is apparently induced by the in-chain CDW correlations. The effect of the CuO chain CDW state on the ZF-$\mu$SR signal was clearly demons
well correlated with the changes in the Cu(2) NQR spectrum with temperature if the assumption of \( \Delta \) being \( T \)-independent is lifted. Figure 1 shows a comparison of fits of the ZF-\( \mu \)SR spectra of YBa\(_2\)Cu\(_3\)O\(_{6.985}\) with \( \Delta \) as a \( T \)-independent parameter to fits achieved with \( \Delta \) free to vary with \( T \). As in the study by Zhang et al., the \( T \)-independent value of \( \Delta \) was obtained by a global fit of ZF-\( \mu \)SR spectra recorded over the full temperature range. Note that the deviation of the fits with a \( T \)-independent \( \Delta \) from the measured ZF-\( \mu \)SR signal becomes apparent beyond 6 \( \mu \)s, necessitating an analysis of the spectra to longer times.

Figure 2(a) shows a comparison of the temperature dependence of \( \lambda_{2F} \) in YBa\(_2\)Cu\(_3\)O\(_{6.95}\) from Ref. 1 to results obtained from fitting the ZF-\( \mu \)SR signals of YBa\(_2\)Cu\(_3\)O\(_{6.985}\) from Ref. 3 with \( \Delta \) as a \( T \)-independent fit parameter. The features below 100 K are enhanced in the higher doped sample and as shown in Fig. 2(b) are well correlated with the changes in the the Cu(2) NQR linewidth measured in YBa\(_2\)Cu\(_3\)O\(_7\) (Ref. 2). The small broad peak argued to occur at \( T^* \sim 77 \) K by Zhang et al. is in fact not a peak, rather there is a dip in \( \lambda_{2F} \) near 60 K that occurs on an otherwise increased or increasing \( \lambda_{2F} \) below 100 K. This is evident from the NQR data, which show no feature near 77 K, but instead exhibit a local maximum near 60 K. As explained in Ref. 2, other kinds of experiments provide evidence for an unbuckling of the CuO\(_2\) layers near 60 K. Even in the absence of CDW order, such a structural change modifies the nuclear dipole contribution to the ZF-\( \mu \)SR signal by changing the distance between the \( \mu^+ \) and the host nuclei. Consequently, the effect is a modification of the relaxation rate near 60 K, which has been observed over a wide doping range from underdoped YBa\(_2\)Cu\(_3\)O\(_{6.50}\) through to overdoped (Ca-doped) samples 2,3,7.

A comparison of the results of similar \( T \)-independent \( \Delta \) fits of the ZF-\( \mu \)SR spectra for YBa\(_2\)Cu\(_3\)O\(_{6.92}\) to the YBa\(_2\)Cu\(_3\)O\(_{6.95}\) data of Zhang et al. is shown in Fig. 3. The good agreement between these independent measurements indicates that sample quality and/or experimental factors are not relevant to the interpretation.

**FIG. 1.** (Color online) Fits of representative ZF-\( \mu \)SR spectra for YBa\(_2\)Cu\(_3\)O\(_{6.985}\) assuming (a)-(c) \( \Delta \) is \( T \)-independent as in Ref. 3, and (d)-(f) \( \Delta \) varies with \( T \) as in Ref. 3.

**FIG. 2.** (Color online) Results of fits to Eq. (2) assuming \( \Delta \) is \( T \)-independent. (a) Comparison of the temperature dependence of the exponential relaxation rate \( \lambda \) in YBa\(_2\)Cu\(_3\)O\(_{6.95}\) from Ref. 1 to \( \lambda \) obtained from fits of the YBa\(_2\)Cu\(_3\)O\(_{6.985}\) ZF-\( \mu \)SR spectra reported in Ref. 3. (b) Comparison of the temperature dependence of \( \lambda \) in YBa\(_2\)Cu\(_3\)O\(_{6.985}\) to the temperature dependence of the Cu(2) NQR linewidth in YBa\(_2\)Cu\(_3\)O\(_7\) reported in Ref. 3.
of the ZF relaxation data. Again, the slight maximum of $\lambda_{ZF}$ near 77 K is not due to critical slowing down of magnetic fluctuations, but rather an artifact of assuming the nuclear dipole contribution to the ZF-$\mu$SR signal is described by a $T$-independent static Gaussian KT function. As for the case of YBa$_2$Cu$_3$O$_{6.95}$, better fits are achieved with $\Delta$ free to vary with temperature.

Zhang et al. also claim to have measured a small peak in the ZF relaxation rate of YBa$_2$Cu$_3$O$_{6.72}$ and a small broad peak in the LF relaxation rate of YBa$_2$Cu$_3$O$_{6.77}$ (in weak LF) near $T^*$ at 210 K and 160 K, respectively.

At 160 K the muon is on the verge of diffusing and is clearly diffusing at 210 K, as established in measurements of YBa$_2$Cu$_3$O$_{6.67}$ in Ref. 3. The width of the internal field distribution associated with the nuclear dipoles in YBa$_2$Cu$_3$O$_y$ is sufficiently narrow that the muon spin polarization in ZF decreases over the full time range of the recorded ZF-$\mu$SR spectra (i.e. up to 14 $\mu$s). Slow muon diffusion reduces the polarization decay at later times, but with increased $\mu^+$ hopping rate $\nu$ the ZF-$\mu$SR signal evolves into an exponential function with a relaxation rate that decreases with increasing $\nu$. In other words, the effect of muon diffusion on the ZF-$\mu$SR signal of YBa$_2$Cu$_3$O$_y$ is a reduction of the relaxation rate. As shown in Fig. 2(a) of Ref. 3, $\lambda_{ZF}$ in YBa$_2$Cu$_3$O$_{6.72}$ decreases above $T \sim 160$ K. A decrease of $\lambda_{ZF}$ above 160 K is also evident in Fig. 2(b) here, where the Cu(2) NQR linewidth is $T$-independent. The small maximum in $\lambda_{ZF}$ observed in YBa$_2$Cu$_3$O$_{6.72}$ near 210 K could originate from the mobile $\mu^+$ reaching and becoming trapped by defects during its short lifetime. The important point here is that the internal fields sensed by the muon have a time dependence above $T \sim 160$ K due to muon diffusion, which renders the $\mu^+$ an ineffective probe of critical magnetic fluctuations at higher temperatures.

When a static LF much greater than the nuclear dipole field distribution is applied, the muon spin decouples from the nuclear dipoles such that the nuclear contribution to the relaxation vanishes. However, when the muon is diffusing it may experience fluctuating field components transverse to the applied static LF that cause an increase in the LF relaxation rate. The reported maximum in $\lambda_{LF}$ near 160 K in YBa$_2$Cu$_3$O$_{6.77}$ (Fig. 2(b) in Ref. 3) may then result from the onset of muon diffusion and subsequent trapping by defects at higher temperatures. It is important though to point out that the peaks claimed in the relaxation rates of the underdoped samples are extremely small ($\lesssim 0.01 \mu$s$^{-1}$), near the reliable detection limit of the method and on the order of statistical jumps in the data. Hence their very existence is questionable.

Finally, from LF-$\mu$SR measurements just above $T_c$ in fields $0.002 \leq B_{1,LF} \leq 0.35$ T, Zhang et al. conclude that there are fluctuating magnetic fields in the pseudogap regime with fluctuation rates as expected for ordered loop currents. The correlation times and rms local field sensed by the muon are determined by fitting LF scans just above $T_c$ to the Redfield formula (see Fig. 1 in Ref. 3). Yet the presented $\lambda_{LF}$ versus $B_{1,LF}$ data clearly deviate from the Redfield formula. Consequently, the quantitative information obtained from these fits is invalid. While there does appear to be a gradual reduction of $\lambda_{LF}$ with increasing $B_{1,LF}$ for two of the three data sets, the values and variation of $\lambda_{LF}$ are so small (on the order of $10^{-3} \mu$s$^{-1}$) that extrinsic effects cannot be ruled out. For example, the incoming positive muons experience magnetic field components perpendicular to their momentum from the fringe fields at the end of the magnet used to generate $B_{1,LF}$. This modifies the muon beam focusing. A gradual reduction of $\lambda_{LF}$ with increasing $B_{1,LF}$ may then originate from a small increase in the fraction of muons that miss the sample. The size of this effect will depend on the sample size and radial position of the sample in the muon beam. Because the LF relaxation rates are below the typical reliable detection limit, the control experiments performed by Zhang et al. on pure Ag can only rule out this scenario if the precise sample size, shape and position are replicated.

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