The origin of the PG in high transition temperature ($T_c$) cuprate superconductors is an enduring mystery, but is widely believed to be a manifestation of a thermodynamic transition to a phase that breaks various symmetries. The apparent detection of an ubiquitous IUC magnetic order in various cuprate families by polarized neutron diffraction $[1, 2]$ lends support to a longstanding idea that electronic loop-current order with translation-invariant quasi-static internal magnetic field of electronic origin in the superconducting (SC) and pseudogap (PG) phases. In both samples the internal magnetic field persists up to 160 K, but muon diffusion prevents following the evolution of the field to higher temperatures. We consider the evidence from our measurements in support of PG order parameter candidates, namely, electronic loop currents and magnetoelectric quadrupoles.

We report muon spin relaxation (μSR) measurements of optimally-doped and overdoped Bi$_{2+x}$Sr$_{2-x}$CaCu$_2$O$_{8+δ}$ (Bi2212) single crystals that reveal the presence of a weak temperature-dependent quasi-static internal magnetic field of electronic origin in the superconducting (SC) and pseudogap (PG) phases. In both samples the internal magnetic field persists up to 160 K, but muon diffusion prevents following the evolution of the field to higher temperatures. We consider the evidence from our measurements in support of PG order parameter candidates, namely, electronic loop currents and magnetoelectric quadrupoles.

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muon hop rate, or equivalently the average rate at which there are changes in the local magnetic field sensed by the muon \( G_{\text{KF}} \). The temperature dependence of the fitted muon hop rate \( \nu \) with a zero offset correction is shown in Fig. 1(b), together with earlier results for underdoped Y123 \[13\]. The zero offset of \( \nu \) suggests the magnetic field distribution in Bi2212 is not as close to a Gaussian as in Y123. Such deviations from a Gaussian form are to be anticipated, have been observed in La214 \[21\], and can be quantitatively accounted for if the muon site(s) and electric field gradients in the material are known. With the zero offset correction, the hop rate in Bi2212 obeys an Arrhenius relationship similar to Y123. The temperature dependence of the fit in Fig. 2(a) shows a significant increase with decreasing temperature with \( \Delta \) as a common parameter yield \( \Delta = 0.0958(6) \mu s^{-1} \) and \( \Delta = 0.095(1) \mu s^{-1} \) for the OD80 and OP91 samples, respectively. Figure 2 shows the temperature dependence of \( \lambda_{\text{ZF}} \). For comparison, the inset of Fig. 2(a) shows \( \lambda_{\text{ZF}} \) versus \( T \) measured in a sample of 99.998 % pure Ag comparable in size to the Bi2212 samples. The ZF relaxation rate in Ag is solely due to weak nuclear dipole fields, and is essentially negligible. However, a \( T \)-dependent relaxation rate can arise from thermal contraction of the sample holder, which slightly changes the position of the sample in the three orthogonal pairs of Helmholtz coils used to cancel the external magnetic field. The results here imply that the thermal energy required to break the O-\( \mu \) bond in Bi2212 is comparable to Y123. Unfortunately, muon diffusion masks clear evidence of PG magnetic order above 160 K.

The ZF-\( \mu \)SR asymmetry spectra are well described by the product of an exponential relaxation function and a static Gaussian ZF Kubo-Toyabe function intended to account for the nuclear-dipolar contribution. Specifically,

\[
G_z(t) = G_{\text{KT}}(\Delta, t) \exp(-\lambda_{\text{ZF}} t).
\] (1)

Global fits of the ZF-\( \mu \)SR spectra as a function of temperature with \( \Delta \) as a common parameter yield \( \Delta = 0.0958(6) \mu s^{-1} \) and \( \Delta = 0.095(1) \mu s^{-1} \) for the OD80 and OP91 samples, respectively. Figure 2 shows the temperature dependence of \( \lambda_{\text{ZF}} \). For comparison, the inset of Fig. 2(a) shows \( \lambda_{\text{ZF}} \) versus \( T \) measured in a sample of 99.998 % pure Ag comparable in size to the Bi2212 samples. The ZF relaxation rate in Ag is solely due to weak nuclear dipole fields, and is essentially negligible. However, a \( T \)-dependent relaxation rate can arise from thermal contraction of the sample holder, which slightly changes the position of the sample in the three orthogonal pairs of Helmholtz coils used to cancel the external magnetic field. The results on Ag show that any such change in \( \lambda_{\text{ZF}} \) between 184 K and 10 K is less than 0.0014 \( \mu s^{-1} \). On the other hand, \( \lambda_{\text{ZF}} \) in the Bi2212 samples exhibit a significant increase with decreasing \( T \) below 160 K. Such behavior reflects a change in the linewidth of the internal magnetic field distribution sensed by the muon ensemble, which cannot be explained in terms of muon diffusion alone. This is our main finding.

The values of \( T^* \) for Bi2212 are ill defined. The temperature ranges for \( T^* \) indicated in Fig. 2 come from a compilation of values measured by different techniques \[31\]. Because the range of experimental values for \( T^* \) extend above 160 K, we cannot say whether there is a spontaneous ZF relaxation appearing at the PG onset.

To determine whether the local magnetic field detected in the PG region is static or fluctuating, we performed LF-\( \mu \)SR measurements on each sample just above \( T_c \),
FIG. 2. (Color online) Temperature dependence of the exponential ZF relaxation rate in the (a) OP91 and (b) OD80 samples. The two data sets for the OD80 sample are from measurements performed during different beam periods. The range of values for the PG temperature $T^*$ are from Ref. [31]. The inset in (a) shows the ZF relaxation rate measured separately in a $5.45 \times 6.54 \times 0.25$ mm sample of 99.998% pure Ag foil. The values of $\lambda_{ZF}$ for Ag come from fits to a single exponential relaxation function.

Theory expression [33]

$$\lambda_{LF} = \frac{\gamma_\mu^2 \langle B^2_\mu \rangle \tau}{1 + (\gamma_\mu B_{LF} \tau)'^2},$$

where $\gamma_\mu/2\pi$ is the muon gyromagnetic ratio, $\langle B^2_\mu \rangle$ is the mean of the square of the fluctuating transverse field components, and $1/\tau$ is the characteristic fluctuation rate of the local fields $B_\mu$. Fits to this equation yield $\langle B^2_\mu \rangle^{1/2} = 1.4(2)$ G and $1/\tau = 0.7(1)$ MHz for the OP91 sample, and $\langle B^2_\mu \rangle^{1/2} = 1.3(3)$ G and $1/\tau = 0.6(2)$ MHz for the OD80 sample. Thus the residual local internal magnetic field sensed by the muon is quasi-static and on the order of the resultant field of the nuclear dipoles. In ortho-II Y123, NMR measurements place upper limits of 4 G and 0.3 G for any magnetic field fluctuating slower than $\sim 0.01$ MHz at the planar and apical oxygen sites, respectively [34]. A similar upper bound for static local fields at the apical oxygen site has been deduced from NMR on HgBa$_2$CuO$_{4+\delta}$ (Hg1201) [35]. Hence, the weak quasi-static fields detected here in Bi2212 are likely fluctuating too fast to be detected by NMR.

Below 160 K where the implanted muon is immobile, the $T$-dependent $\lambda_{ZF}$ may originate from a continuous
change in the nuclear dipole contribution or be caused by magnetic dipole moments of electronic origin. The former may result from structural changes that modify the distance between the muon and nuclear spins, as well as the direction of the maximal local electric field gradient (EFG) that defines the quantization axis for the nuclear spins. A \( T \)-dependent electric quadrupolar interaction of the nuclei with the local EFG can also result from a gradual development of charge inhomogeneity or charge order. While \( ^{17}\text{O} \) NMR measurements on overdoped Bi2212 (\( T_c = 82 \text{ K} \)) demonstrate an inhomogeneous distribution of local EFG at the O(1) sites in the CuO\(_2\) plane, this does not evolve with temperature \( \text{[36]} \). X-ray scattering measurements on underdoped Bi2212 show the development of CDW order within the PG phase \( \text{[37]} \), persisting as weak dynamic CDW correlations near \( T^* \) \( \text{[38]} \). Indeed, short-range CDW order has been identified in recent years to be ubiquitous in cuprates \( \text{[39]} \). However, the CDW correlations are most pronounced in the underdoped regime and significantly weaken or fade away near optimal doping. Moreover, in contrast to \( \lambda_{\text{ZF}} \) (Fig. 2) the CDW correlations are suppressed below \( T_c \). Hence CDW correlations do not seem to explain the \( T \)-dependent ZF relaxation rate observed below 160 K.

Another potential source of the ZF relaxation is dilute magnetic impurities. Dilute remnants of the underdoped phase containing Cu spin correlations fluctuating slow enough to be detectable on the \( \mu\text{SR} \) time scale are unlikely to be present near and above optimal doping. Bulk magnetization measurements down to 2 K show no evidence of a magnetic impurity or secondary phase. As shown in Fig. 3, the normal-state magnetic susceptibility of the OP91 sample measured up to 300 K exhibits no low-\( T \) upturn indicative of trace amounts of a paramagnetic impurity.

A previous weak LF-\( \mu\text{SR} \) study of overdoped polycrystalline samples of Bi\(_2\)Sr\(_2\)Ca\(_{1-x}\)Y\(_x\)Cu\(_2\)O\(_{8+\delta}\) in a LF of 20 G revealed a small increasing relaxation rate below \( \sim 135 \text{ K} \) in a \( T_c = 81 \text{ K} \) sample \( \text{[40]} \). While attributed to an inhomogeneous distribution of internal magnetic field generated by SC domains, the frequency scale for SC fluctuations above \( T_c \) (\( 10^{10} \) to \( 10^{14} \) Hz) established by other methods \( \text{[41]-[43]} \) is too high to produce an observable LF relaxation. Inserting \( \langle B^{2}_{\mu} \rangle^{1/2} \leq 20 \text{ G} \) and \( 1/\tau = 10^{10} \text{ Hz} \) in Eq. (3) yields \( \lambda_{\text{LF}} \lesssim 3 \times 10^{-4} \text{ } \mu\text{s}^{-1} \), which is far smaller that the LF relaxation rates reported in Ref. \( \text{[40]} \) and well below the reliable detection limit. There is strong evidence for fluctuating SC domains in Bi2212 and Y123 well above \( T_c \) from high transverse-field (TF) \( \mu\text{SR} \) experiments, which are highly sensitive to a distribution of time-averaged internal magnetic fields \( \text{[44]} \). Nonetheless, SC domains do not affect the ZF relaxation rate.

The IUC magnetic order in Bi2212 inferred by neutrons is characterized by a pair of staggered magnetic moments in the CuO\(_2\) plane predominantly perpendicular to the CuO\(_2\) plane and displaced from a Cu atom along the [1, 1, 0] direction, with an ordered magnetic moment of \( \sim 0.1 \mu\text{B} \) \( \text{[8]} \). While the precise muon site in Bi2212 is undetermined, the muon is expected to reside near an O atom. Calculations of the magnetic dipolar field generated by \( \text{static} \) IUC magnetic order at the oxygen sites in the CuO\(_2\), SrO and BiO planes yield 3.1 G, 116 G and 1.4 G, respectively. Two of these values are on the order of the magnitude of the quasi-static internal field just above \( T_c \) estimated from the LF-\( \mu\text{SR} \) data. However, this does not exclude the possibility of magnetic order fluctuating too fast to be detectable on the \( \mu\text{SR} \) time scale, or equivalently \( \gamma^2_{\mu} \langle B^2_{\mu} \rangle \tau \lesssim 0.001 \text{ } \mu\text{s}^{-1} \). With this said, the detected field is close to the calculated size of the internal magnetic fields generated by \( \text{quasi-static} \) magnetoelectric quadrupolar ordering — estimated to be \( \sim 0.3 \text{ G} \) at the oxygen sites in Hg1201 \( \text{[29]} \) and potentially larger at the muon site(s) in Bi2212.

In summary, we have detected by \( \mu\text{SR} \) a weak \( T \)-dependent \( \text{quasi-static} \) internal magnetic field in the SC and PG phases of Bi2212, seemingly of electronic and intrinsic origin. While consistent with a static version of the IUC magnetic order inferred from neutrons measurements, this interpretation is difficult to reconcile with \( \mu\text{SR} \) studies of other cuprates. Our findings offer some support for a theory ascribing the primary order parameter in the PG phase to quasi-static magnetoelectric quadrupoles.

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