The origin of the pseudogap region below a temperature $T^*$ is at the heart of the mysteries of cuprate high-temperature superconductors. Unusual properties of the pseudogap phase, such as broken time-reversal and inversion symmetry are observed in several symmetry-sensitive experiments: polarized neutron diffraction, optical birefringence, dichroic angle-resolved photoemission spectroscopy, second harmonic generation, and polar Kerr effect. These properties suggest that the pseudogap region is a genuine thermodynamic phase and are predicted by theories invoking ordered loop currents.

Recent experiments have shown that, consistent with a phase transition, time-reversal symmetry ($5, 6$) and spatial rotation and inversion symmetries ($7–9$) are broken below $T^*$ in a number of cuprate superconductors. However, lattice translational symmetry is preserved, indicating that the antiferromagnetism that lies near the pseudogap phase in the temperature-doping phase diagram is not involved. The unbroken translational symmetry has focused attention on the phenomena within the crystalline unit cell. In particular, the broken time-reversal symmetry is predicted by theories that invoke states with intra-unit-cell broken translational symmetry has focused attention on the phenomena near the pseudogap.

INTRODUCTION

The mysterious pseudogap phase in high-temperature superconducting cuprates has been the subject of an enormous amount of research ($1–4$) but with little consensus on either its origin or its role in high-temperature superconductivity. The name arises from the loss of low-lying electronic excitations below a temperature $T^*$, which depends strongly on the hole concentration on CuO$_2$ planes. This loss results in considerable modification of properties connected with these excitations, but their nature has been difficult to determine. Even whether or not $T^*$ is in fact a phase transition transition temperature has been controversial.

Recent experiments have shown that, consistent with a phase transition, time-reversal symmetry ($5, 6$) and spatial rotation and inversion symmetries ($7–9$) are broken below $T^*$ in a number of cuprate superconductors. However, lattice translational symmetry is preserved, indicating that the antiferromagnetism that lies near the pseudogap phase in the temperature-doping phase diagram is not involved. The unbroken translational symmetry has focused attention on the phenomena within the crystalline unit cell. In particular, the broken time-reversal symmetry is predicted by theories that invoke states with intra-unit-cell (IUC) magnetic order ($10–12$). However, doubt has been cast on the existence of IUC magnetic order: Local static magnetic fields of the expected magnitude were not observed in muon spin rotation ($\mu$SR) ($13–18$) or nuclear magnetic resonance (NMR) ($19, 20$) experiments. $\mu$SR and NMR are magnetic resonance techniques in which spin probes (implanted muons or nuclei) are sensitive to magnetic behavior on the local (atomic) scale. $\mu$SR in particular is capable of detecting nearby static magnetic moments in the range of 0.001 to 0.01 $\mu_B$ and did not do so in cuprate systems where neutron diffraction measurements observed moments of the order of 0.1 $\mu_B$.

IUC magnetic order would be retained, and the absent static field would be accounted for, if the ordered moments fluctuate among alternate orientations ($15, 18, 19$). This would average the local field $B_{loc}(t)$ at spin-probe sites to zero for time scales longer than a characteristic correlation time $\tau_c$. These fluctuations could be due to finite-size domains of an ordered phase with different field orientations ($21$), as seen in tunneling spectroscopy ($22$). For NMR and $\mu$SR, the experimental time scale for this averaging is considerably longer ($\geq 10^5$ s) than that for the other techniques ($\leq 10^3$ s). Thus, all experiments would be consistent if the fluctuations were “slow,” with $\tau_c$ between these limits.

Averaging or "motional narrowing" of $B_{loc}(t)$ (the term comes from the effect of nuclear motion on static NMR line broadening in an applied field) occurs in the rapid fluctuation limit $\gamma B_{loc}^{\text{rms}} \tau_c \ll 1$, where $\gamma$ is the gyromagnetic ratio of the nucleus or muon and $B_{loc}^{\text{rms}} = \langle B_{loc}(t)^2 \rangle^{1/2}$ is the root-mean-square (rms) local field. For the muon, $\gamma = 8.5156 \times 10^8$ s$^{-1}$ T$^{-1}$. The IUC-ordered moments per triangular plaquette obtained from neutron diffraction [$0.05 \text{ to } 0.1 \mu_B$ in Y$_2$Ba$_2$Cu$_3$O$_6$+$\delta$ ($5, 6$) and progressively lower at higher $y$] give dipolar field values $B_{loc} = 1$ to $10$ mT at candidate muon sites in the unit cell of La$_2$–xSr$_x$CuO$_4$ ($15$). For these fields, the above inequality is satisfied for $\tau_c \approx 10^{-6}$ s, which is in the range of experimental consistency. Thus, $B_{loc}(t)$ gives rise to dynamic or "spin-lattice" nuclear or muon spin relaxation. Itoh et al. ($23$) have reported “ultraslow” fluctuations in HgBa$_2$CaCu$_2$O$_{6+\delta}$, that may be of this kind.

In $\mu$SR experiments, the motionally narrowed dynamic muon spin relaxation rate in zero applied field ($ZF$) is given by $\lambda_{ZF} = (\gamma B_{loc}^{\text{rms}})^2 \tau_c$. The muon $\mu$SR experiments have shown that the absence of static fields expected for magnetic order. This may be due to other mechanisms such as the formation of magnetic domains with finite size.
The relaxation rate measured in a longitudinal magnetic field (LF) (that is, the magnetic field parallel to the initial muon polarization) depends on the magnitude \( H_L \) of the field, an effect of sweeping the muon on Zeeman frequency through the fluctuation noise power spectrum \((24, 25)\). For Markovian fluctuations with a single well-defined correlation time \( \tau_c \), the LF relaxation rate \( \lambda_{LF} \) in a field \( H_L \) is given by the so-called Redfield relation \((24)\)

\[
\lambda_{LF}(H_L) = \frac{\gamma_{\mu}^2 (B_{rms}^{loc})^2 \tau_c}{1 + (\gamma_{\mu} H_L \tau_c)^2}
\]  

(1)

The dependence of \( \lambda_{LF} \) on \( H_L \), if observed to be of the form of Eq. 1, provides estimates of \( \tau_c \) and \( B_{rms}^{loc} \). More generally, one expects a decrease of \( \lambda_{LF} \) for \( \gamma_{\mu} H_L \) greater than a characteristic fluctuation rate \( 1/\tau_c \), in which case \( \tau_c \) and \( B_{rms}^{loc} \) from fits of Eq. 1 to the data are heuristic estimates of the characteristic time and field scales. The Redfield relation has been widely applied in \( \mu \)SR to characterize dynamic fluctuating magnetic fields in strongly correlated electron systems, for example, in the heavy fermion superconductor \( \text{PrOs}_4\text{Sb}_{12} \) \((26)\).

Here, we report the discovery of slow magnetic fluctuations in single crystals of \( \text{YBa}_2\text{Cu}_3\text{O}_y \), \( y = 6.72, 6.77, 6.83 \), and 6.95 (superconducting transition temperatures \( T_c = 72, 80, 88, \) and 91 K, respectively) via the field dependence of the \( \mu \)SR dynamic relaxation. Consistency with Eq. 1 is found, and values of \( \tau_c \) and \( B_{rms}^{loc} \) are obtained. We also find maxima at \( T_{mag} \approx T^* \) in the temperature dependences of the rates in ZF and LF. This is consistent with critical slowing down of magnetic fluctuations near the transition and demonstrates that these fluctuations are associated with the IUC order. There is considerable statistical uncertainty because the measured relaxation rates are near the limit of the technique, but standard statistical analysis techniques (see below) demonstrate the validity of our conclusions.

**RESULTS**

We performed LF-\( \mu \)SR experiments on samples with \( y = 6.72, 6.77, \) and 6.83. The field dependence of the exponential relaxation rate \( \lambda_{LF} \) was measured for these samples at temperatures below \( T^* \) and above \( T_c \) for \( 2 \text{ mT} \leq \mu_{\mu} H_L \leq \text{400 mT} \). The minimum field was chosen to be much larger than the \( \sim 0.1\text{-mT} \) quasi-static field from nuclear dipoles (see Materials and Methods and section S1), so that the dipolar field is “decoupled” \((25)\); that is, the resultant field is nearly parallel to the muon spin, and the dipolar fields do not cause appreciable muon precession. Thus, one observes only dynamic relaxation.

The results are shown in Fig. 1, together with fits of Eq. 1 to the data. The rates are very small, close to the lower limit accessible to the technique, and the statistical uncertainty is large. Control experiments and precautions taken to minimize systematic errors are discussed in section S2.

In Fig. 1 (A and B), the data lie above the fits for \( H_L \leq \text{5 mT} \). This might suggest a logarithmic field dependence as an empirical description of the data. However, there is no clear physical mechanism for this in \( \text{YBa}_2\text{Cu}_3\text{O}_y \). In highly anisotropic tetracyanoquinodimethane (TCNQ) charge transfer salts, nuclear spin relaxation due to diffusion of electronic spin quasi-particles exhibits log \( H \) behavior for particular values of the anisotropic hopping rates \((27)\). However, there is no reason to suspect this diffusive conduction in cuprates, and the higher low-field rates in Fig. 1 are more likely to be due to incomplete decoupling for these fields.

Table 1 gives values of \( \tau_c \) and \( B_{rms}^{loc} \) from the fits, together with standard deviation (SD) (1 \( \sigma \)) statistical uncertainties. The experimental values of \( B_{rms}^{loc} \) differ from zero by 5 to 9 \( \sigma \) individually and \( \sim 10 \sigma \) cumulatively; nonzero values are established at this level. Both \( \tau_c \) and \( B_{rms}^{loc} \) vary smoothly with \( y \). We carried out dipolar lattice sum calculations for \( B_{rms}^{loc} \) assuming candidate muon stopping sites from the literature \((28, 29)\) and approximating the current loops as point dipoles \((30)\). These yield estimates \( B_{rms}^{loc} \approx \text{1 to 1.5 mT} \) for \( 0.1\mu_{\mu} \) loop-current magnetic moments, of the same order of magnitude as the observed values. The calculated values are not changed significantly for the “criss-cross” bilayer loop-current configurations recently reported by Mangin-Thro et al. \((31)\).

It is evident that \( \tau_c \) falls in the middle of the range of experimental consistency discussed above. The observed increase of \( \tau_c \) with increasing \( y \) could be due to the approach to a quantum critical point as \( T_{mag} \rightarrow \text{0} \). However, fluctuations of the short-range IUC magnetic order may be associated with defects \((21)\), in which case \( \tau_c \) could depend on sample preparation and not be an intrinsic property. More work is required to elucidate the nature of the observed fluctuations.

Exponential relaxation is observed in ZF together with the expected Gaussian contribution due to random quasi-static dipolar fields from nuclear moments (compare section S1). Just above \( T_c \), \( \lambda_{ZF} \) for \( y = 6.72 \) (Fig. 2A) is a factor of about 5 higher than \( \lambda_{LF} \) above \( T_c \) (Fig. 1A). Some of this increase is due to a Lorentzian contribution to the distribution of static fields \((13)\), but some of it is doubtless because of dynamic relaxation; in ZF, the two are hard to disentangle experimentally (section S1).

The temperature dependence of \( \lambda \) was measured in ZF and LF \((H_L = \text{4 mT})\) for various samples. Figure 2 (A and B) shows \( \lambda_{ZF}(T) \) and \( \lambda_{LF}(T) \), respectively, for \( y = 6.72 \). Fig. 2C shows \( \lambda_{LF}(T) \) for \( y = 6.77 \), and Fig. 2D...
DISCUSSION

The observed increase of motionally narrowed muon spin relaxation with decreasing temperature below \(T^*\), shown in Fig. 2, cannot occur in a transition to a uniformly ordered state. It is, however, consistent with low-frequency fluctuations in domains of IUC magnetic order, of increasing magnitude with the increasing order parameter \(n\). Scanning tunneling spectroscopy experiments (22) have found these domains in the pseudogap phase associated with defects, with linear dimensions of \(\sim 20\) unit cells; these provide a mechanism for the pseudogap in the one-particle spectra (21). Other experiments (38, 39) have observed mysterious anomalous low-frequency fluctuations in the pseudogap phase ascribed to extended defects.

Phenomena other than IUC magnetism can affect \(\mu\)SR experiments. Sonier and co-workers (14, 16) observed features in ZF data from \(YBa_2Cu_3O_y\) and attributed them to charge and structural inhomogeneities from lattice changes and CDW formation. Fits of an exponentially damped Gaussian Kubo-Toyabe (KT) to data from a nearly fully doped sample \((y = 6.985)\) (14) yielded correlations between \(\Delta(T)\) and \(\lambda(T)\), leading to the conclusion that the exponential rate could not be determined unambiguously (40). Structure in ZF \(\lambda(T)\) with \(\Delta(T)\) held fixed was related to the onset of short-range CDW order (41) for that doping.

For slightly lower doping \((y = 6.95)\), however, fits with \(\Delta(T)\) a free parameter (42) yield a clear peak in \(\lambda(T)\) at \(T_{\text{mag}} = 80\) K with no corresponding structure in \(\Delta(T)\) at that temperature (compare section S3). NQR measurements (43) on a sample of similar oxygen content showed that CDW order sets in at a considerably lower temperature \((\sim 35\text{ K})\). For \(y < 6.95\), ZF CDW transition temperatures from NMR/NQR and other measurements are significantly lower than \(T_{\text{mag}}\) from our experiments. For all \(y\), the onset of long-range CDW order is at still lower temperatures and is only observed in high-applied magnetic fields (20). Recent torque magnetometry measurements also provide strong evidence that CDW order and the pseudogap phase are characterized by distinct broken symmetries at different temperatures (9). The detailed temperature-doping phase diagram (Fig. 3) shows that lattice change/CDW and pseudogap phases set in at different temperatures and are distinct phenomena. We conclude that there is no evidence for associating the maxima in \(\lambda(T)\) and its increase at low temperatures with either CDW or charge inhomogeneity.

It has been claimed (40) that the maxima for \(y = 6.72\) and 6.77 at \(\sim 210\) and \(\sim 165\text{ K}\), respectively (Fig. 2, A and C), could be due to the onset of thermally activated muon hopping (diffusion) (14); this causes unwanted dynamic relaxation by nuclear dipolar fields. However, in the usual trapping-detrapping scenario (44), the maximum is due to a temperature sequence in which the muons are trapped and static at low temperatures, begin hopping with increasing temperature, find deep impurity traps and become static again at an intermediate temperature, and finally detract definitively at high temperatures. It has been argued (42) that this is highly unlikely in the present case because independent \(\mu\)SR determinations (14, 42, 45) of the muon hopping rate using dynamical Gaussian KT fits (25) show that hopping is too slow to produce the observed decrease in \(\lambda(T)\) below the maximum (compare section S4).

Alternatively, the maximum might be purely dynamic because with increasing temperature, the muon hopping rate passes through the muon Zeeman frequency. In that case, however, the application of \(H_{\text{L}} = 4\text{ mT}\) field would move the maximum to temperatures where the hopping rate is \(\sim -\gamma_\mu H_{\text{L}} = 3.4\text{ }\mu\text{s}^{-1}\). According to the dynamic KT fits, this
is well above 300 K, whereas in our data (Fig. 2B), the position of the maximum is unchanged. We conclude that muon hopping is not the origin of the maximum for \( y = 6.72 \). Note that, for \( y = 6.67 \), no maximum in \( \lambda_{ZS}(T) \) was seen near \( T^* \approx 200 \) K (13), which is probably due to significant muon hopping at this temperature.

Previous transverse-field (TF) \( \mu\text{SR} \) experiments in the pseudogap phase (46) observed exponential relaxation and ascribed it to static spatial inhomogeneity of superconducting fluctuations. Our observed LF-\( \mu\text{SR} \) rates (Fig. 1) are an order of magnitude slower than the TF-\( \mu\text{SR} \) rates. This is consistent with the assumption that the latter are static and precludes detecting the dynamic relaxation in TF-\( \mu\text{SR} \).

The observed critical slowing down at \( T_{\text{mag}} \approx T^* \) (Fig. 2) indicates that this temperature marks the onset of broken time-reversal symmetry, as is also found in each of the four hole-doped cuprate families amenable so far to polarized neutron diffraction experiments. The observed magnitude of the order parameter of about 0.1 for \( \lambda_{ZS}(T) \) was seen near \( T^* \approx 200 \) K (13), which is probably due to significant muon hopping at this temperature.

Our discovery of fluctuating magnetic fields provides an understanding of the absence of static magnetic fields due to IUC magnetic order in \( \text{YBa}_2\text{Cu}_3\text{O}_y \). The expected fields are present but fluctuating. Although \( \mu\text{SR} \) is a point probe in real space and, thus, is not directly sensitive to the spatial symmetry, our results are strong evidence for IUC order and its excitations and establish them as important for understanding the unusual behavior of cuprates.

**MATERIALS AND METHODS**

**Sample growth and characterization**

High-quality single crystals of \( \text{YBa}_2\text{Cu}_3\text{O}_y \) were grown by the top-seeded solution growth polythermal method using \( \text{BaO-5CuO} \) solvent (48). A \( \text{YBa}_2\text{Cu}_3\text{O}_y \) single crystal with an \( ab \) plane area of \( 10 \text{ mm} \times 10 \text{ mm} \) and \( c \) axis length of \( 8 \text{ mm} \) was synthesized with a cooling rate of 0.5 K per hour in air. The crystal was then cut into small pellets with thicknesses of 0.55 mm and lateral dimensions of \( 2 \text{ mm} \times 2 \text{ mm} \). Single crystals with optimal \( T_c = 91 \) K were achieved by annealing at 400°C for 180 hours in flowing oxygen. A range of oxygen concentrations of \( \text{YBa}_2\text{Cu}_3\text{O}_y \) was achieved by post-annealing in flowing oxygen at different temperatures as described in the study by Gao et al. (49), resulting in superconducting transition temperatures between 72 and 88 K.

Figure 4 shows the temperature dependences of the magnetization and the resistivity in our samples. The data indicate that the superconducting transitions are sharp. The values of \( T^* \) from the departure of the resistivity from linearity are in agreement with previous results (50).

**Muon spin relaxation experiments**

In the time differential \( \mu\text{SR} \) technique (51), spin-polarized muons are implanted into the sample. In the muon decay to a positron and two neutrinos, the positron momentum is preferentially oriented along the direction of the muon spin at the time of decay. The time evolution of the ensemble muon polarization can therefore be monitored via the asymmetry in positron emission count rates.

Our \( \mu\text{SR} \) experiments were performed using the Los Alamos Meson Physics Facility spectrometer at the M20 beamline of TRIUMF (Vancouver, Canada) and the MUSR and EMU spectrometers at ISIS, Rutherford...
The inverse relative SDs (IRSDs) (the N in “Nσ”) for $B_{\text{loc}}^{\text{rms}}$ (Table 1) and for the maxima in the temperature dependences at $T_{\text{mag}}$ (Fig. 3) are shown in Table 2. The IRSD for $B_{\text{loc}}^{\text{rms}}$ is simply its value divided by its SD. For the maxima at $T_{\text{mag}}$, baseline points were chosen above and below each maximum, and baseline values at intermediate points were estimated by linear interpolation. The IRSD of each point is its amplitude relative to the baseline divided by its SD, and the IRSD of the maximum is the square root of the sum of squares of the IRSDs of the points. The sign of the amplitude was included in this sum to account for negative contributions. For both $B_{\text{loc}}^{\text{rms}}$ and the maxima, the cumulative IRSD is the square root of the sum of squares of the individual sample IRSDs. It can be seen that some individual IRSDs are marginal, but the cumulative values are quite satisfactory.

### Table 2. IRSDs of $B_{\text{loc}}^{\text{rms}}$ and relaxation rate maxima near $T_{\text{mag}}$ from muon spin relaxation rates in YBa$_2$Cu$_3$O$_y$.

| $y$   | $B_{\text{loc}}^{\text{rms}}$ | $H_\text{loc}$ (mT) | No. of points | IRSD |
|-------|----------------|-----------------|---------------|------|
| 6.72  | 4.8            | 0               | 7             | 4.5  |
| 6.72  | 4              | 11              | 3.8           |
| 6.77  | 8.7            | 4               | 15            | 5.2  |
| 6.83  | 6.2            | 0               | 3.9           |
| 6.95  | 6.2            | 6               | 3.9           |
| Cumulative | 11.7         | 0               | 8.8           |

Random error in μSR experiments arises from the Poisson distribution of positron count rates. The MUSRFIT analysis software computes the propagation of this error to that of the parameters of the fit function and reports their probable SD $σ$. All parameter uncertainties quoted in this article are $1σ$.

### Statistical analysis

Random error in μSR experiments arises from the Poisson distribution of positron count rates. The MUSRFIT analysis software computes the propagation of this error to that of the parameters of the fit function and reports their probable SD $σ$. All parameter uncertainties quoted in this article are $1σ$.

### SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/4/eaao5235/DC1

section S1. Muon relaxation functions

section S2. Control experiments

section S3. Temperature dependence of static Gaussian KT relaxation rate $Λ_{\text{KT}}(T)$

section S4. Muon hopping, superconductivity

section S5. Superconducting fluctuations

section S6. High-temperature relaxation

fig. S1. ZF relaxation of the muon asymmetry $A_i(t)$ in YBa$_2$Cu$_3$O$_{6.72}$

fig. S2. Time evolution of the positron count rate asymmetry $A_i$ at various temperatures and fields in single-crystal YBa$_2$Cu$_3$O$_y$

fig. S3. LF muon spin relaxation rates in silver samples.

fig. S4. Fits of representative ZF-μSR spectra from YBa$_2$Cu$_3$O$_{6.76}$ for $Δ_{\text{ff}}$ fixed and free.

fig. S5. Temperature dependence of ZF exponential damping rate $λ_{\text{ex}}$ and static Gaussian KT rate $Λ_{\text{KT}}$ for YBa$_2$Cu$_3$O$_{6.72}$

fig. S6. Damped dynamic Gaussian KT fit of ZF data from YBa$_2$Cu$_3$O$_{6.72}$

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