Hole-doped perovskite manganites of the form La$_{1-x}$Ca$_x$MnO$_3$ (LCMO) feature a ferromagnetic metallic ground state for 0.2 < $x$ < 0.5, with the highest $T_C$ for the combined ferromagnetic and metal-insulator transition at an optimal doping of $x \approx 0.3$. The colossal magnetoresistance (CMR) observed at this transition cannot be explained solely by Zener double-exchange; rather, the physics underlying the CMR effect in LCMO likely arises from strong coupling between the spin, charge, lattice, and orbital degrees of freedom and nanoscale inhomogeneities between the spin, charge, lattice, and orbital degrees of freedom and nanoscale inhomogeneities between competing phases. For $x = 0.3$ LCMO, short-range static and dynamic polaron correlations are observed above $T_C$, with an ordering wave vector of (0.25 0.25 0) signifying CE-type charge- and orbital-ordered regions. The spin dynamics in the ferromagnetic phase are likewise unconventional. The spin-wave dispersion softens near the zone boundary, which can be fit to a phenomenological model of first- and fourth-nearest-neighbor ferromagnetic Heisenberg interactions, and displays anomalous spin-wave damping. The spin-wave stiffness coefficient renormalizes but does not fully collapse as $T_C$ is approached from below. This contrasts with higher-bandwidth manganite materials such as Pr$_{1-x}$Sr$_x$MnO$_3$ where the stiffness fully collapses at $T_C$ as expected for a second-order ferromagnetic phase transition. Above $T \approx 200$ K a spin diffusive quasielastic component develops in the low-$q$ spectral weight arising from a short-ranged localization of electrons on the Mn$^{4+}$/Mn$^{3+}$ lattice. This quasielastic component displays a short field dependence and dominates the spin fluctuation spectrum near $T_C$ with a temperature dependence that closely matches those of both lattice polarons and the bulk resistivity. It is the development of this spin diffusive component, rather than the thermal population of spin waves, that truncates the ferromagnetic metallic ground state in a weakly first-order phase transition.

In this work we report neutron spectroscopy measurements on a single crystal sample of La$_{0.7}$Ca$_{0.3}$MnO$_3$. Spin correlations at 265 K, slightly above $T_C$, are explored primarily through the $Q$ dependence of scattering at constant energy transfer. The long-wavelength spin dynamics can be described by spin diffusion with a short, almost temperature-independent correlation length of $\approx 12$ Å. For $q$ transfers approaching the Brillouin zone edge an additional anisotropic scattering component is observed at low energies, in the form of ridges of strong quasielastic scattering running along $(H 0 0)$ and equivalent directions. Well-defined $Q$-space correlations are observed in constant-$E$ scans at energies up to at least 28 meV, suggesting robust short-range spin correlations in the paramagnetic phase.
The $q$ position corresponding to the maximum intensity of the ring increases with the energy value of the constant-$E$ scan consistent with the $\omega \propto q^{-2.5}$ expectation of dynamical scaling theory; particularly, the ring radii, as measured in scans along the $(\xi 0 0)$ direction, are consistent with a quasielastic half width at half maximum (HWHM) that varies with $q$ as $\Gamma(q) = \Delta q^{-2.5}$ where $\Delta = 18.9 \pm 0.5$ meV Å$^{-2.5}$. Closer to the zone boundary, the isotropy of the spin dynamics breaks down and ridges of scattering are present which connect the rings along $(H 0 0)$ and equivalent directions. These ridges of scattering are strongest in Brillouin zones at low $Q$ and the intensity falls off at higher $Q$ in a manner roughly consistent with the Mn form factor squared. Uncertainties throughout this paper are statistical and refer to one standard deviation.

These data reflect the integrated intensity of transverse scans through scattering ridges, with the energy integrated over $2 \text{meV} \leq h\omega \leq 6 \text{meV}$. The red line is the Mn form factor squared. In the higher-$Q$ data, such as the $3.5$ SrO data, such as the $P_{0.5}(Ca_{0.8}Sr_{0.2})_{0.45}MnO_3$ signifying short-range antiferromagnetic correlations. In $La_{0.7}Cr_{0.3}MnO_3$, the anomalous scattering near the $(0.5 0 0)$ position instead arises from a breakdown in dynamical scaling theory in which the energy width of the quasielastic scattering ceases to be isotropic as $q$ approaches the zone boundary, with smaller quasielastic widths for $q$ values along the $(\xi 0 0)$ direction.

The intensity of this scattering anisotropy is energy-dependent, as shown in the constant-$Q$ energy scans of Fig. 3(a). For smaller $q$ values where the data are well described by simple spin diffusion, such as the $q = 0.325$ Å$^{-1}$ data shown in the figure, the energy scans do not depend on the orientation of $q$. In the higher-$q$ data, such as the $q = 0.812$ Å$^{-1}$ data shown in the figure, the energy of the ridge of extra scattering is demonstrated by the difference in scattering for the $q \parallel (\xi 0 0)$ and $q \parallel (\xi -\xi 0)$ data. Figure 3(b) shows this difference in scattering intensity for $q = 0.649$ Å$^{-1}$; this $q$ position is shifted slightly from the center of the ridge as the zone edge position will feature a nuclear superlattice reflection in the elastic data. This anomalous scattering near the zone edge is quasielastic in nature, having a maximum at the elastic position and an energy HWHM of approximately $2.5$ meV. This anisotropy ceases to be measurable for $h\omega \gtrsim 15$ meV; this is roughly the energy transfer where rings of scattering surrounding adjacent Brillouin zone centers begin to overlap. While this intensity is energy-dependent, the transverse width
FIG. 3. (Color online) (a) Energy dependence of the 265 K paramagnetic scattering in constant-$Q$ scans. $Q$ values have been chosen so that $\vec{q}$ away from (0 1 0) is along either the ($\xi$ 0 0) or ($\xi$ −$\xi$ 0) directions, and with the magnitude of $q$ either 0.325 Å$^{-1}$ or 0.812 Å$^{-1}$. The data have been folded across the $K = 0$ plane and integrated over ±0.13 Å$^{-1}$ in all $\vec{q}$ directions. (b) The energy dependence of the ridge intensity, displayed as the difference in scattering for measurements at $Q = (0.6 \ 0)$ and (0.283 0.717 0) (both with $q = 0.649$ Å$^{-1}$). The gray line is a guide to the eye.

The presence of persistent spin correlations at 265 K can be further explored through the spin dynamics in the ($H$ $K$ 0.5) scattering plane. For low-energy transfers, the spin correlations in this plane display a more complex structure. To increase statistics the measured data have been folded across the $L = 0$ plane, so that data with $L = −0.5$ and $L = 0.5$ have been averaged together. The intensity plot of correlations in the ($H$ $K$ 0.5) plane, shown as Fig. 5(a), is qualitatively quite similar to the correlations in the ($H$ $K$ 0) plane at lower energies. A similar anisotropy between scans along the ($\xi$ 0 0) and ($\xi$ −$\xi$ 0) directions is observed, as shown in Fig. 5(b); these ridges have a transverse width, shown in Fig. 5(c), consistent with the calculated spin-wave scattering.
so that all correlations in this plane are in the high-$q$ regime where simple spin diffusion theory should not be applicable. In particular, the correlations yield peaks in $q$ that are far narrower than would be expected from a purely diffusive model. A breakdown in scaling theory in which the width of peaks in constant-$E$ scans falls far below the theoretical value at high $q$ was also observed in paramagnetic iron and nickel. These correlations are also qualitatively similar to the ferromagnetic phase $Q$-space spin-wave correlations displayed in LCMO. The bilayer manganite $La_{1.3}Sr_{1.8}MnO_7$ was likewise reported to display $Q$-space spin correlations in the paramagnetic phase that qualitatively resembled those of ferromagnetic spin waves. Given the dispersion relation displayed by propagating spin waves in LCMO below $T_C$, the $Q$-space correlations in the $(H K 0)$ plane at an energy of $\omega_0$ will have the same structure as correlations in the $(H K 0.5)$ plane at an energy of $\omega_0 + \Delta_0$ where $\Delta_0 = 4S|J_1|$ (such that $\Delta_0 \approx 25$ meV at 100 K). Finding the correlations of Fig. 5 in data where $\hbar\omega \approx 26$ meV suggests a significant renormalization of $\Delta_0$ from the 100 K data; this is reminiscent of the previously reported renormalization of the spin-wave stiffness, where $D(T_C) \approx D(T = 0)/2$.

It is clear that the spin correlations in LCMO above $T_C$ result in well-defined peaks in constant-$E$ scans that qualitatively resemble the correlations from the spin-wave excitations below $T_C$; similar results have been previously reported in a bilayer manganite. Despite these well-defined peaks in constant-$E$ scans, no clear peaks at finite energy are observed in constant-$Q$ scans. For $\vec{q}$ positions away from the $(H 0 0)$ direction, the positions of these peaks are well described by spin diffusive dynamical scaling theory. The data at higher-$q$ values deviate from the expectations of dynamical scaling theory primarily through peaks in the $Q$-space correlations that are far narrower than the simple spin diffusive model would predict. An additional component of the paramagnetic scattering is also observed as ridges of unexpectedly strong quasielastic scattering at low-energy transfers and $\vec{q}$ positions parallel to $a^*$ or a symmetry equivalent direction. Intrinsic inhomogeneity with small scale regions of competing phases is a common signature in strongly correlated electron materials, including stripe order in high-$T_C$ superconducting cuprates and polar nanoregions in relaxor ferroelectrics. Phase separation of this sort is well known in CMR manganites, with considerable evidence for lattice polarons and spin polarons; LCMO samples with smaller doping levels also display evidence of ferromagnetic droplets. The physics of colossal magnetoresistance in LCMO has been modeled as a percolation or Griffiths phase effect arising from the separation of various competing phases.

The effects of hole doping and applied field on the spin fluctuation spectrum of LCMO near $T_C$ suggest that the small-$q$ spin diffusion portion of the scattering arises from the short length-scale hopping of electrons on the Mn$^{3+}/Mn^{4+}$ lattice; this diffusive scattering coexists with spin waves near $T_C$ and drives the ferromagnetic phase transition. It is also known that the temperature dependence of the low-$q$ quasielastic scattering is quite similar to the temperature dependences of the bulk resistivity and the polaron correlations, suggesting a close connection between paramagnetic scattering and colossal magnetoresistance. The new high-$q$ quasielastic scattering in the paramagnetic phase described in this work represents spin correlations with wavelengths approaching atomic length scales, comparable in size to the small polarons generated by localized electrons. We hope that further measurements on the short-range spin correlations near $T_C$ will shed new light on the physics of colossal magnetoresistance.

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