Spin dynamics of the quasi-two-dimensional ferromagnetic bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$

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We have investigated the spin dynamics of the quasi-two-dimensional bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$, which shows colossal magnetoresistance (CMR) behavior, by inelastic neutron scattering. We have taken advantage of the recently developed multiplexing technique with a three-axis spectrometer to map simultaneously a large region of the (Q,ω) space in a low-dimensional magnetic system. We have measured the spin wave dispersion at $T=16$ K along [100], followed the temperature evolution of the spin wave dispersion close to $T_C \approx 128$ K and also measured the scattered neutron intensities in the (Q,ω) space above T_C up to $T=200$ K. The magnetic inelastic intensity above T_C looks qualitatively very similar to those below T_C and have a structure in the (Q,ω) space suggesting persistent spin waves. However, although there exist well defined peaks at finite energies in constant energy slices up to the highest temperature investigated, no well defined peaks at finite energy are observed above T_C in constant-Q slices of the spin response measured in the (Q,ω) space.

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The discovery of colossal magnetoresistance (CMR) in the bilayer compound $La_{2-2x}Sr_{1+2x}Mn_2O_7$ (Ref. 1) has generated a lot of investigations focussed on understanding the microscopic mechanism of this phenomenon. Due to its reduced dimensionality, the electronic and magnetic properties of this bilayered manganite are expected to be different from those for the well-studied three-dimensional manganite $La_{1-x}A_xMnO_3$ (A=Sr, Ca, Ba). Indeed, some novel features have already been reported.[2–5] The reduced dimensionality in fact enhances the CMR effect, albeit at the cost of decreasing the ferromagnetic transition temperature. Further, some features concern the observation of the coexistence of ferromagnetic and antiferromagnetic short-range-ordered spin correlations[3,6] above $T_C \approx 126$ K. Osborn *et al.*[5] have observed canting of the magnetic moments above T_c. We reported[6–8] inelastic-neutron-scattering investigations of the magnetic excitations in $La_{1.2}Sr_{1.8}Mn_2O_7$ leading to a complete determination the exchange interactions at low temperature and also the temperature dependence of the spin-wave excitations. We further reported[9] the results of the investigation of diffuse magnetic scattering above T_C up to room temperature. The spin dynamics of the bilayer manganites have also been investigated by other groups.[10–14] Despite an early claim by Perring *et al.*[15] that nearest-neighbor localized Heisenberg model can account for the spin dynamics of perovskitelike ferromagnetic CMR manganites, more careful work on perovskitelike manganites, as well as on bilayer manganites[7,8,10,11,16–21] revealed that the spin dynamics of ferromagnetic manganites cannot be described by a localized Heisenberg model in the whole Brillouin zone. The dominant experimental signatures are the spin-wave softening and damping observed especially close to the zone boundary.

In CMR manganites there are two species of electrons which contribute to the low-energy physics: Mn t_{2g} electrons form the localized spins with high-spin state $S=3/2$, whereas itinerant electrons occupy the Mn e_g bands. A simple model which captures the physics of CMR ferromagnetic manganites is that of the double-exchange (DE) Hamiltonian first introduced by Zener.[22] Zener proposed that, in order to gain maximum kinetic energy, the hopping (t_{ij}) of the itinerant Mn e_g electrons, which are held parallel to the localized t_{2g} electrons by Hund's coupling (J_H), will cause the localized spins to align in a parallel way leading to the ferromagnetic ground state. The magnetic excitation of the double exchange model has been discussed by Furukawa[23,24] who showed that for a strong Hund's coupling $J_H \to \infty$ and in the classical spin limit $S \to \infty$, the spin waves in a DE model can be mapped into an effective Heisenberg model. Shannon and co-workers[19,20] have discussed the quantum and thermal corrections of the minimal DE model and have shown that a part of the experimentally observed zone-boundary softening and damping can be qualitatively accounted for by these corrections. In order to explain quantitatively the spin-wave softening and damping effects, it may be necessary to include orbital and lattice degrees of freedom to the minimum DE

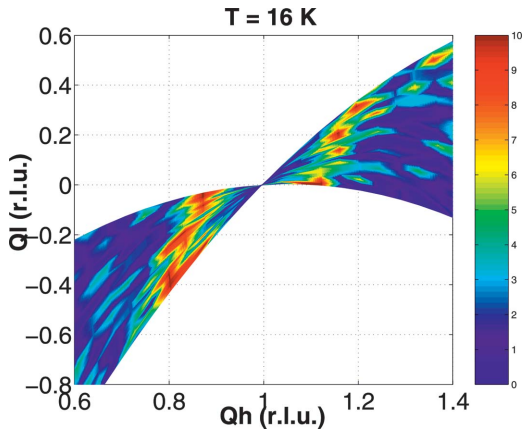

FIG. 1. (Color) The covered reciprocal space points of $La_{1.2}Sr_{1.8}Mn_2O_7$ during an energy scan. The movements in the c^* direction are quite small and do not influence the disperison significantly within statistics.

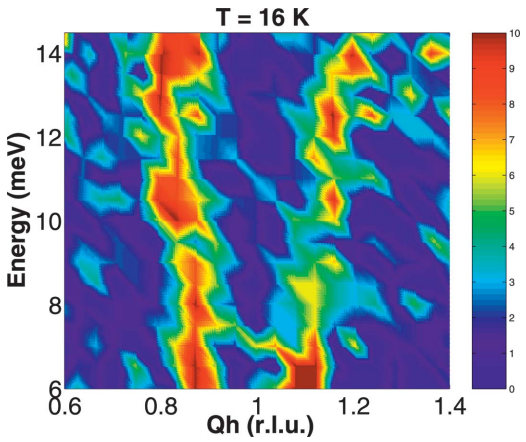

FIG. 2. (Color) Mapping of the spin-wave dispersion in $La_{1.2}Sr_{1.8}Mn_2O_7$ in a single energy scan at $T=16$ K in the energy transfer range 6-14 meV by using the multianalyzer detector (MAD). The dispersion of the spin wave is clearly seen.

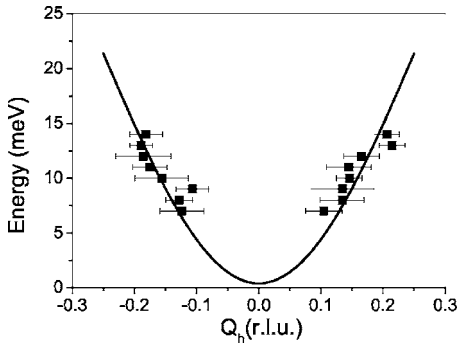

FIG. 3. Dispersion of the acoustic spin waves in $La_{1.2}Sr_{1.8}Mn_2O_7$ at $T=16$ K obtained by fitting the slices of the $(\mathbf{Q}_0,\omega_0)$ plane of Fig. 2 at several constant energies, which are equivalent to the so-called constant-E scans of a three-axis spectrometer with a single detector.

Hamiltonian. The above theoretical conclusions are equally valid for a perovskitelike 3D as well as quasi-2D manganites, such as bilayer $La_{1.2}Sr_{1.8}Mn_2O_7$.

In order to explore further the validity and shortcomings of the minimum DE model experimentally, we have investigated the temperature dependence of the spin dynamics of the CMR bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$. The recently developed multianalyzer detector (MAD) for a thermal three-axis spectrometer at the Institut Laue-Langevin and also the quasi-2D nature of $La_{1.2}Sr_{1.8}Mn_2O_7$ have enabled us to map the scattered neutron intensities in the $(\mathbf{Q},\omega)$ space during a single scan. The same large cylindrical single crystal of $La_{1.2}Sr_{1.8}Mn_2O_7$ of diameter 5- and 25-mm long of our previous investigations[6–9] was used for the present study.

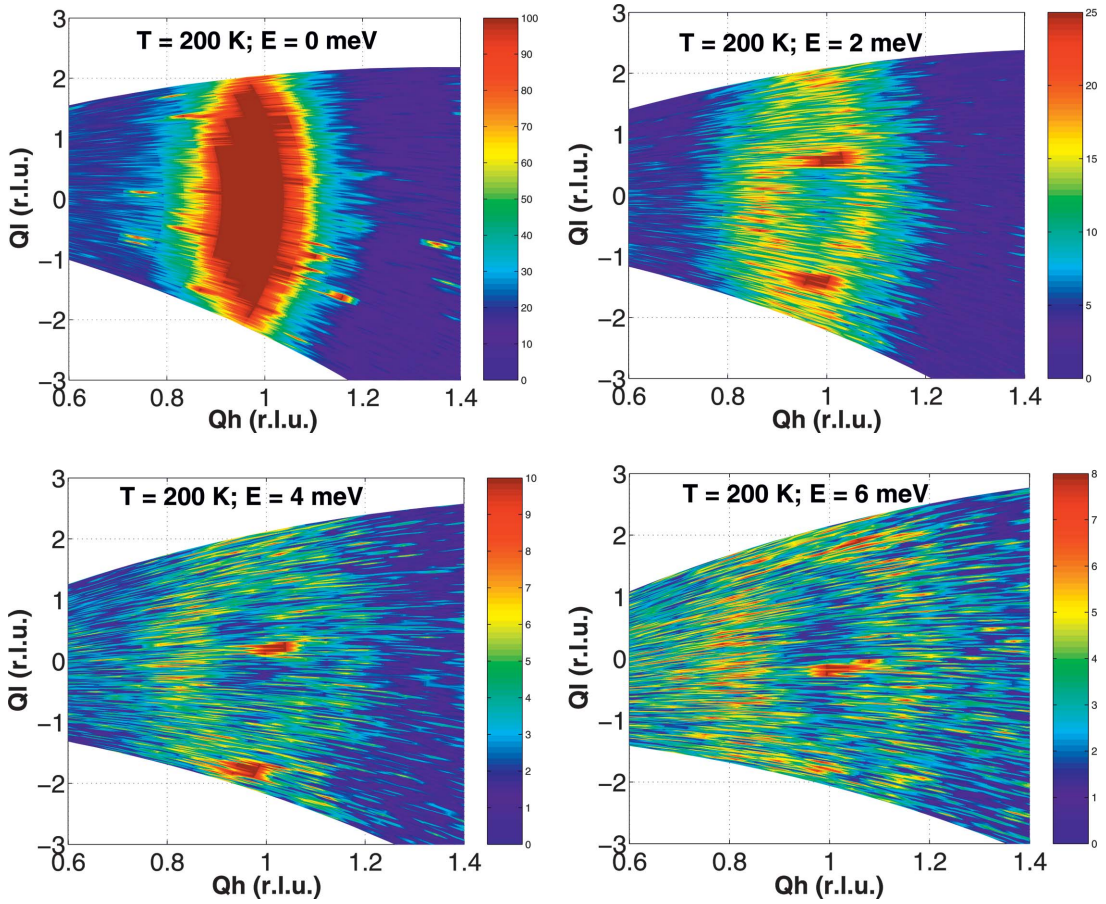

FIG. 4. (Color) Energy analysis of the diffuse modulated rod-like scattering from $La_{1.2}Sr_{1.8}Mn_2O_7$ at $T=200$ K for energy transfers of $E=0$, 2, 4 and 6 meV.

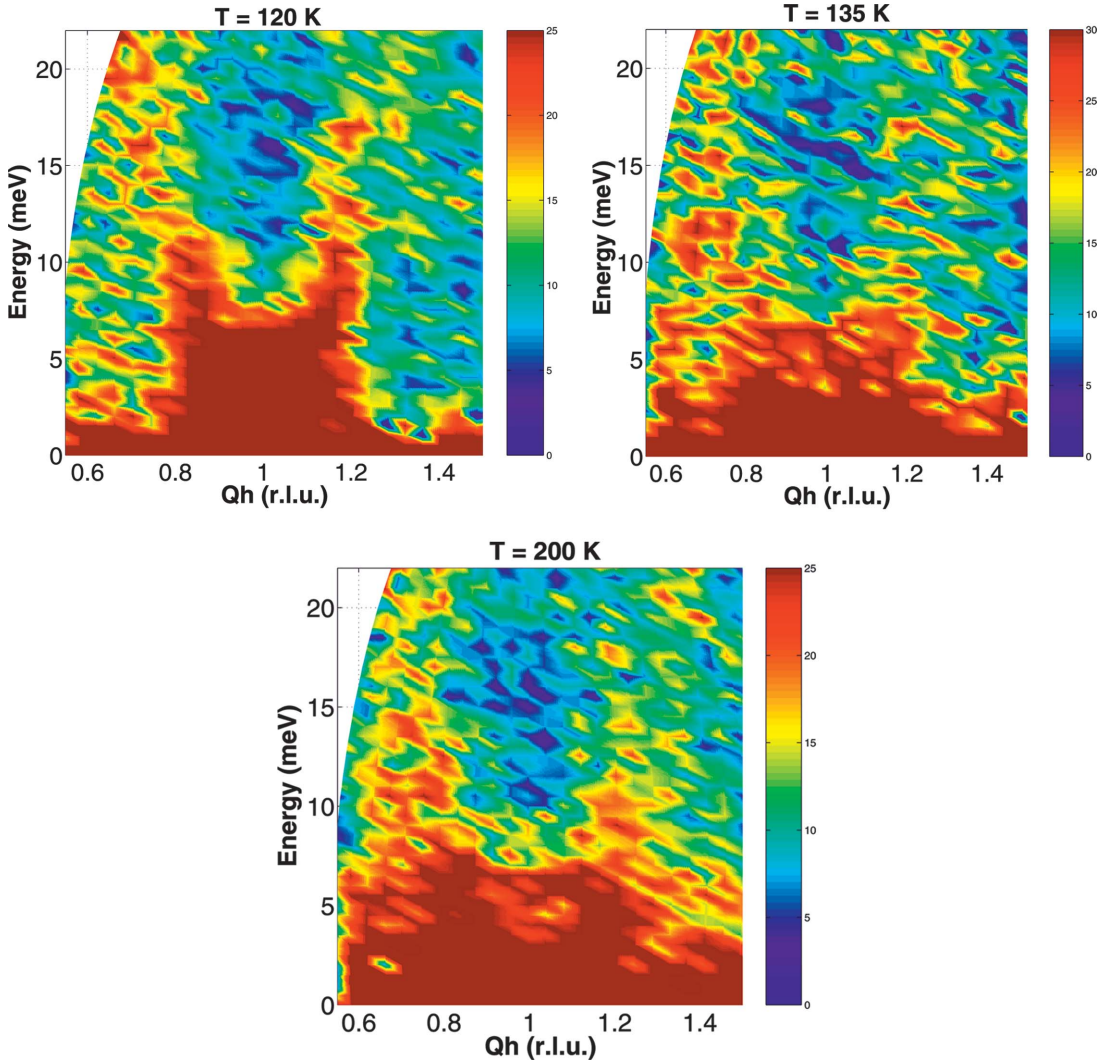

FIG. 5. (Color) Mapping of the spin wave dispersion in $La_{1.2}Sr_{1.8}Mn_2O_7$ at $T=120$, 135, and 200 K by using the multianalyzer detector (MAD).

Inelastic-neutron-scattering experiments were performed on the thermal three-axis spectrometer IN3 installed at the thermal guide at the Institut Laue-Langevin in Grenoble. The crystal was put inside a helium cryostat with its [010] crystallographic axis vertical so that the scattering plane was $(h0l)$. As a secondary spectrometer a multianalyzer detector setup was installed.[25] All analyzers were aligned to the same final energy $E_f=31$ meV. The energy resolution was 1.2 meV and the Q resolution was about ± 0.03 Å^{-1} for each channel. The main advantage of this technique is its use for low-dimensional systems. For such systems a setup of the crystal with the dispersing axis perpendicular to $\mathbf{k}_i$, allows one to perform a $\mathbf{k}_i$ scan with all detectors in a constant-Q manner simultaneously. As $La_{1.2}Sr_{1.8}Mn_2O_7$ is a quasi-2D magnetic system with strong dispersions along [100] and [010] but only a small dispersion along [001], this type of scan is obviously advantageous. Furthermore, due to the body centered bilayer magnetic spin structure of $La_{1.2}Sr_{1.8}Mn_2O_7$, acoustic and optic magnetic spin wave branches have cosine-type variation in the structure factor along the c direction.[6] Because of this, an energy scan where only the incoming energy k_i is varied, is not advisable. Although the dispersion does not change much along the nonsensitive c-axis, the intensity of one branch can be lost with larger energy transfers. In order to map a large part of one magnon excitation branch in one scan it was necessary to perform a type of scan with an as small as possible variation in Q_l in the interesting Q_h region. This could be achieved by fixing one detector in $\vec{Q}$. The neigboring detectors perform then movements in both directions in the $(h0l)$ plane. As an example, Fig. 1 shows the covered reciprocal points during an energy scan in the range $6-14.5$ meV. The movements in the c^* direction are quite small and does not influence the disperison significantly within statistics. Figure 2 shows the dispersion of the acoustic spin wave measured at 16 K in an (Q_h,E) plot. Constant energy slices from these scans have been fitted with a Gaussian function to yield the inelastic energy positions of the acoustic spin waves in $La_{1.2}Sr_{1.8}Mn_2O_7$ at $T=16$ K. The resulting values are plotted in Fig 3. The continuous curve is a fit to a classical dispersion equation for a Heisenberg ferromagnet: $\hbar\omega=\Delta+2SJ_a[1-\cos(2\pi Qa)]$. In the considered small energy

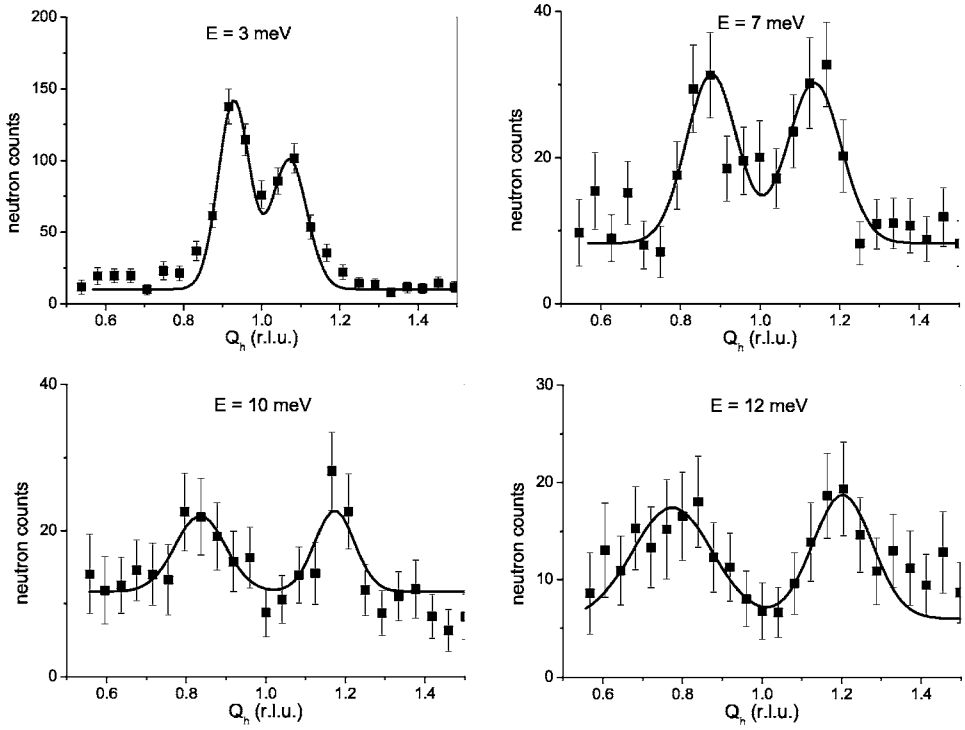

FIG. 6. Constant-E slices of the (Q,ω) mapping of Fig. 5 at $T=120$ K. The continuous curves are least-squares fits with Gaussian functions.

range this equation describes quite well the spin-wave dispersion. The spin value was set to $S=1.8$ and the gap energy was kept constant to $\Delta=0.4$ meV. The resulting value for $J_a=5.8$ meV is quite close to the value obtained from previous measurements using classical three-axis spectrometry, although our measured energy range is quite small.[6–8]

In our previous measurements of the diffuse intensity integrated over energy, modulated rodlike scattering was observed close $Q=(1,0,0)$ at temperatures above T_C up to room temperature.[9] During the present study we measured the scattering from $La_{1.2}Sr_{1.8}Mn_2O_7$ for different energy transfers. Figure 4 shows the rod scattering for energy transfers of $E=0$, 2, 4, and 6 meV at 200 K, well above T_C, whereas in our previous measurements[9] energy-integrated intensities were determined. We see clearly from Fig. 4 that the extended ellipsoidlike rod scattering consists of central essentially elastic magnetic scattering whereas the outer boundary regions are mostly inelastic and dispersive. The hollow ellipsoidlike scattering shape for energy transfers $E>0$ suggested the existence of dispersing inelastic excita-

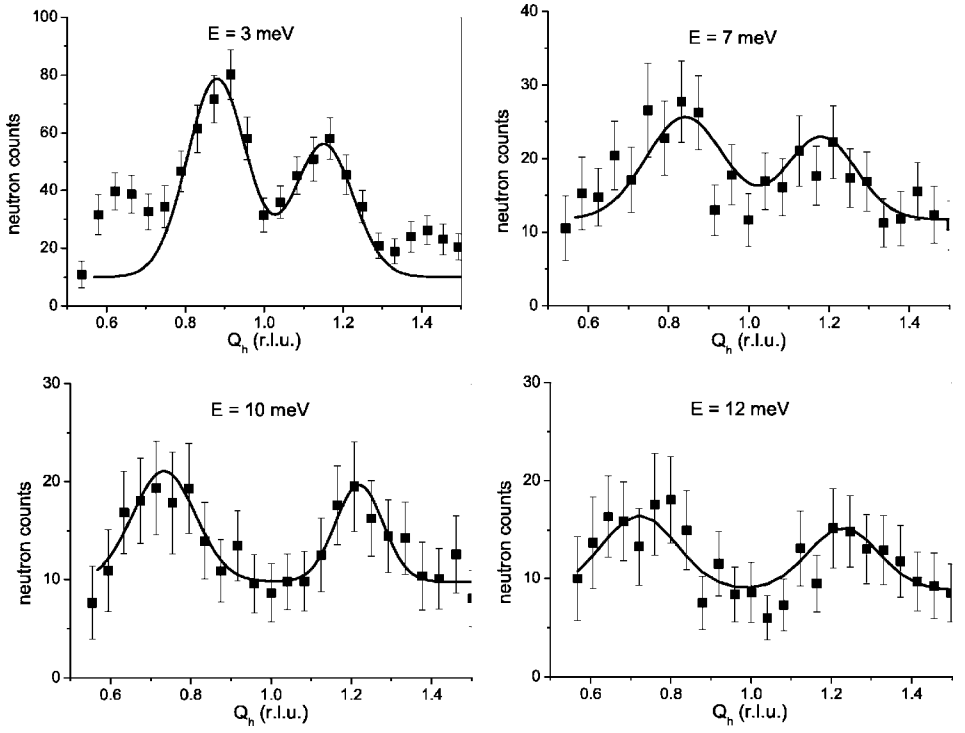

FIG. 7. Constant-E slices of the (Q,ω) mapping of Fig. 5 at $T=200$ K. The continuous curves are least-squares fits with Gaussian functions.

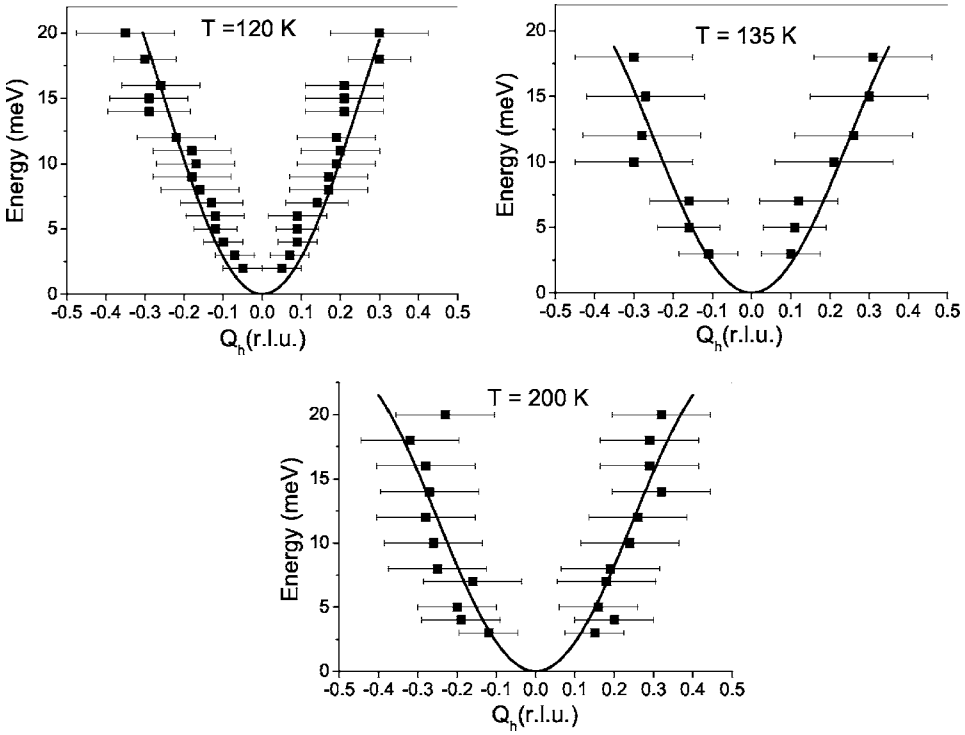

FIG. 8. Spin wave dispersion of $La_{1.2}Sr_{1.8}Mn_2O_7$ at $T=120$, 135, and 200 K. The continuous curves represent fits to the localized Heisenberg model. The data points are obtained from constant-energy cuts of the (Q,ω) maps illustrated in Figs. 6 and 7. The horizontal bars give the widths of the excitations and are not error bars.

tions above T_C. To confirm this we performed energy scans at several temperatures ($T=120$, 135, and 200 K) with the middle detector fixed at $\vec{Q}=(1,0,0)$ as it was already done for the low-temperature scan shown in Fig. 2. Figure 5 shows such energy scans for $La_{1.2}Sr_{1.8}Mn_2O_7$. The dispersive spin-wave-like structure is observed up to temperatures as high as $T=200$ K. Figures 6 and 7 show the constant-energy slices of the (Q,ω) maps of Fig. 5 whereas Fig. 8 shows the corresponding dispersions obtained by fitting Gaussian functions to constant-E slices. The horizontal bars give the width of the fitted curves. Due to the steepness and the width of the excitations a constant-Q slice does not give an inelastic peak anymore. This can be directly seen in the plotted dispersions. Most of the scattered intensity is concentrated close to $E=0$ as can be expected for dominantly diffusive excitations, and what one can deduce already from the constant energy cuts in Fig. 4. Fitting the abovementioned Heisenberg model function to the 120 K data we get a value for the exchange interaction of $J_a=4.1\pm0.16$ meV, which is already softer compared to the low-temperature value. At a first sight the disperion relations above T_C look quite similar to that at low temperature except for the larger widths of the excitations. It is tempting to suppose spin-wave-like excitations also above the transistion temperature. As there is, to our best knowledge, no theoretical model which can describe high energy spin fluctuations in this temeprature range we used the simple function based on the Heisenberg localized model mentioned before also above T_C. The fit for the dispersions at $T=135$ and 200 K gives the same in-plane exchange interaction $J_a=3.3\pm0.2$ meV within the standard deviations. This value is in fact not very different from the result of a modelization of the diffuse scattering rods within a mean field theory at 300 K with $J_a=2.8$ meV.[9] The resultant exchange interaction J_a obtained at several temperatures are summarized in Table I.

The scattering due to spin fluctuations in $La_{1.2}Sr_{1.8}Mn_2O_7$ above T_C may appear unusual at first sight, but this is actually what is expected, at least qualitatively, for a quasi-2D magnetic system such as $La_{1.2}Sr_{1.8}Mn_2O_7$. The intralayer and intrabilayer exchange interactions J_a and J_c are very strong compared to the interbilayer coupling J'.[6–8] These strong exchange interactions make the system quasi-2D and so the two-dimensional bilayers are still short-range ordered above T_C.[9,26] Therefore we suppose that dispersive magnetic excitations in the bilayers can persist above the three-dimensional ordering temperature. Such a behavior is quite typical of localized 2D magnetic systems and has been observed before.[27,28] However, the exact nature of the spin correlations in $La_{1.2}Sr_{1.8}Mn_2O_7$ above T_C remains to be determined. It is not easy to decide whether the spin excitations observed above T_C are heavily damped dispersive spin waves or they merely exhibit a complex diffusive character in the absence of any theoretical calculation of the spin response. Similar controversy over the existence of persistent spin waves in transition metal ferromagnets Fe and Ni has not yet been settled after about three decades of intense research.[29–34] In contrast to $La_{1.2}Sr_{1.8}Mn_2O_7$, these are three-dimensional systems. It is true that although there exist well defined peaks at finite energies in constant energy scans up to

TABLE I. Fitted values of J_a obtained from the dispersions shown in Fig. 8 at different temperatures.

Temperature (K)	J_a (meV)
16	5.8± 0.2
120	4.1± 0.2
135	3.3± 0.3
200	3.3± 0.3

the highest temperature investigated, no well defined peak at finite energy are observed above T_C in constant-Q slices of the spin response. It remains to be checked whether the spin response observed experimentally can be explained by the theoretical spin susceptibilty function calculated on the basis of at least the minimal double exchange model including of course the quasi-two-dimensional character of the magnetic system. The dispersions shown in Fig. 8, which are determined from the Gaussian fit to the constant-E slices of Fig. 7, show a peculiar behavior above T_C. Regarding the dispersion at 200 K one can get the impression that at larger energy tranfers the dispersion is even steeper or is already localized above $Q_h=0.3$. Instead of becoming flat and horizontal at the zone boundary, the two symmetrical branches for q and $-q$ tend to come closer. Whether there is a smooth transition from propagating spin excitations to localized spin fluctuations above T_C is now a matter of investigation. The three-axis spectrometer situated at the neutron guide did not allow us to investigate spin excitations for energy transfers above 20 meV and therefore we can only speculate the dispersion behavior at higher energy transfers.

In conclusion we have mapped the spin response of $La_{1.2}Sr_{1.8}Mn_2O_7$ in a wide temperature range below and above T_C and have discovered unusual spin response above T_C which is neither totally diffusive nor persistent spin-wave-like. Although the constant-Q slices from the (Q,ω) mapping of the spin fluctuations do not reveal any well-defined peaks above T_C, the constant-energy slices do show well-defined peaks. The structure of the (Q,ω) mapping above T_C has a peculiar shape which deserves to be quantitatively explained by model calculations based on a double-exchange model.

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