Griffiths phases vs magnetic polarons in the lightly doped $La_{1-x}Sr_xMno_3$

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During the past two decades, the existence of the Griffith's-phase (GP) regime [1] has been reported in different compounds with complex magnetic interactions such as spin-glasses [2], strongly correlated 4f-electron systems with the non-Fermi liquid behavior [3,4], low-dimensional magnet $CuGeO_3$:Fe [5], perovskite manganites [6–9], and rare-earth intermetallic compound $Tb_5Si_2Ge_2$ [10]. The competition between charge-ordered anti-ferromagnetic (AFM) and metallic ferromagnetic (FM) phases appears to be a significant factor for the rich phase diagrams of these systems.

Despite the large number of works, the nature of nanoscale-size inhomogeneities (stable FM clusters) in the paramagnetic (PM) regime of manganites is still an unresolved problem [11]. Very recently, the existence of Griffith's-like features has been proposed in the PM phase of the $La_{1-x}Sr_xMnO_3$ single crystals for the concentration range of Sr $0.07 \leqslant x \leqslant 0.16$, using electron-spin resonance (ESR) and magnetic susceptibility measurements [9]. These singularities, which are characterized by the coexistence of FM resonance and PM resonance signals above the magnetic ordering temperature T_c and by the deviation of susceptibility from the Curie–Weiss form, were identified as a GP regime. As an example, Fig. 1 shows the resonance positions for $x = 0.1$. The spectrum consists of strongly orientation-dependent resonance lines (in addition to the PM signal due to the majority of Mn^{3+} and Mn^{4+} spins [12] at $g \approx 2$) with unusual g values (max. $g_{eff} \approx 4$). The main PM resonance (solid squares) shifts at the transition to the canted AFM (CA) phase (T_{CA}) and returns to $g \approx 2$ as FM resonance at T_{FM}. At the transition into the FM phase the spectrum changes again, where the second strong resonance (open squares) shifts to high fields due to demagnetization indicating the increase of local magnetic fields in the sample. Remarkably, no such additional FM resonances could be detected above T_c for samples with $x > 0.175$, which already exhibit a FM metallic ground state. Although the co-existence of PM and FM resonances in various manganites was reported previously, an additional FM resonance (solid stars in Fig. 1) in the PM

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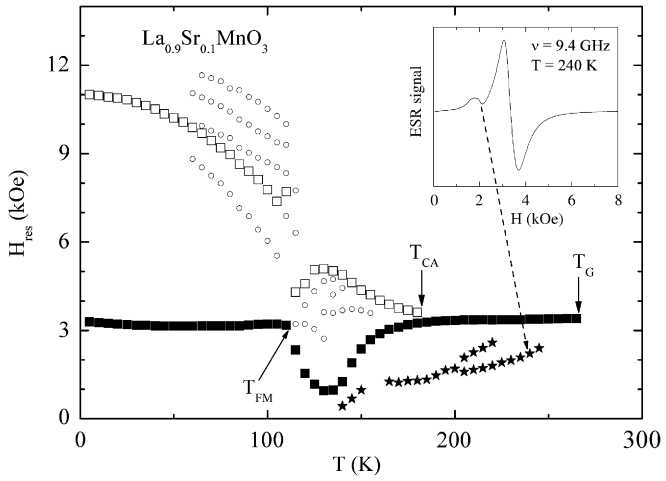

Fig. 1. Temperature dependence of the resonance positions H_{res} in $La_{0.9}Sr_{0.1}MnO_3$: squares: strong resonances; circles: weak resonances; stars: cluster resonance. Inset: ESR spectrum at 240 K.

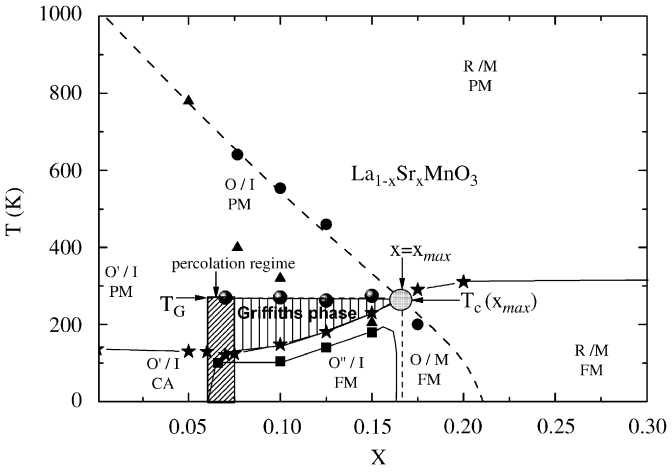

Fig. 2. Phase $T-x$ diagram of $La_{1-x}Sr_xMnO_3$ [9]; dashed = structural and solid = magnetic phase transitions. The intersection (gray circle) of T_G (spheres) with the magnetic boundary T_c (stars) coincides with the transition from the orthorhombic (O) to the rhombohedral (R) structure (I = insulator, M = metal). Data for $x = 0.06$ and 0.07 were taken from Refs. [17] and [18], respectively. Lines are guides to the eye.

regime has been often associated with the persistence of magnetic spin polarons or FM clusters (for example, Refs. [13,14] and references therein).

However, we assume that the origin of these effects can be understood in terms of a GP, which is confined to the restricted triangular region of the $T-x$ phase diagram (Fig. 2), with a Griffith's-like [15] temperature scale $T_G \sim 270$ K above T_c. These effects are arising due to the presence of correlated quenched disorder as a consequence of the transition from the Jahn–Teller (JT) distorted orthorhombic (O') phase into the rhombohedral (R) phase, i.e. as the random locations of the FM bonds begin to fluctuate concomitantly with the fluctuating lattice distortions [9,16]. As a result, the FM clusters are formed thermodynamically during cooling below T_G. The GP regime is limited by the Sr concentration $x_c \sim 0.07$ and the FM transition temperature $T_c(x_{max})$ up to maximal Sr concentration $x_{max} \sim 0.16$. The percolation threshold regime $0.06 < x_c < 0.075$ has been refined by the results of neutron-diffraction studies [17,18]. The observed value of $T_G = 270$ K is naturally explained through the influence of the structural transition, which anneals the disorder distribution at $x \approx 0.16$. For $x > 0.16$, the time scale for disorder fluctuations is no longer much greater than the dynamical scale of the order–parameter fluctuations, and the Griffiths singularities disappear. In this regime (R phase), a description based on magnetic polarons is suitable.

Generally, the polaronic state in manganese perovskites is a consequence of a strong electron–phonon coupling of the JT active Mn^{3+} ions. The hopping of magnetic polarons is widely used to interpret the conductive properties in the PM state of manganites. However, the exact nature of the entity responsible for the ESR line is not yet established. For example, the PM lines at $g \approx 2$ have been ascribed to some complexes consisting of a collection of Mn^{3+} (effective spin $S = 2$) and Mn^{4+} ($S = \frac{3}{2}$) ions coupled by strong exchange interactions [12,19,20]. Two possible structural units (Mn^{4+} or $Mn^{4+}O_4^{7-}$) were proposed for additional resonances with other unusual g values, which were observed in the ESR spectra of the weakly doped $La_{1-x}Sr_xMnO_3$ ($0.05 \leqslant x \leqslant 0.125$) [21]. These centers are autolocalized, but percolate at higher concentrations and lead to conductivity. An analysis of the ESR parameters far above T_{FM} in $La_{0.7}Ca_{0.3}MnO_3$ [22] predicts that the temperature-dependent effective spin is not 2 or $\frac{3}{2}$ per Mn and that the ESR absorption originates from the bottleneck of itinerant electrons and magnetic spin clusters.

In summary, we have demonstrated the importance and the impact of studying the GP in the lightly doped $La_{1-x}Sr_xMnO_3$. For a heaver doping $x > 0.16$, an applicability of polaronic models can be appropriate. Moreover, we propose that the existence of the GP regime may not be restricted to this compound, but rather represents a generic feature of perovskite manganites when a GP becoming observable experimentally due to competition of two phases. We suggest that these observations and the full consequences of our data must be taken into account to test any models being developed to explain the problem of nanoscale phase separation in magnetic solids.

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