Phase Separation or Griffiths Phase in Doped Manganites?

Myron Salamon
University of Illinois

It is quite clear that the colossal magnetoresistance of $La_{2/3}M_{1/3}MnO_3$ involves a percolation like mixture of ferromagnetic/conducting and paramagnetic/resistive regions. Fig. 1, for example, shows the width of EXAFS peaks, related to the distribution of Mn-O bonds. The evolution from a large bond distribution (Jahn-Teller distorted) to smaller bond distribution (undistorted) is obvious. Indeed, we demonstrated some time ago that it is possible to extract the proportions of conducting and insulating regions as functions of magnetic field and temperature directly from the resistivity data. This is shown in Fig.2

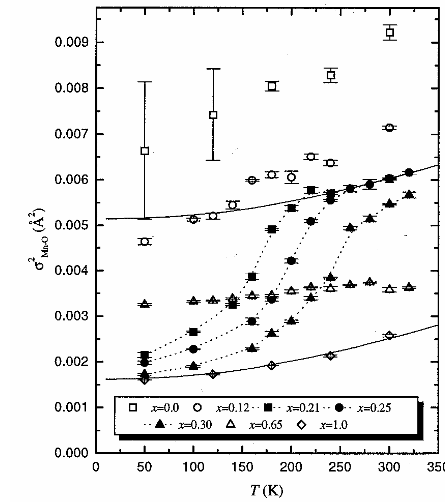


Figure 1. EXFS mean-square bond distribution (Booth et al.)

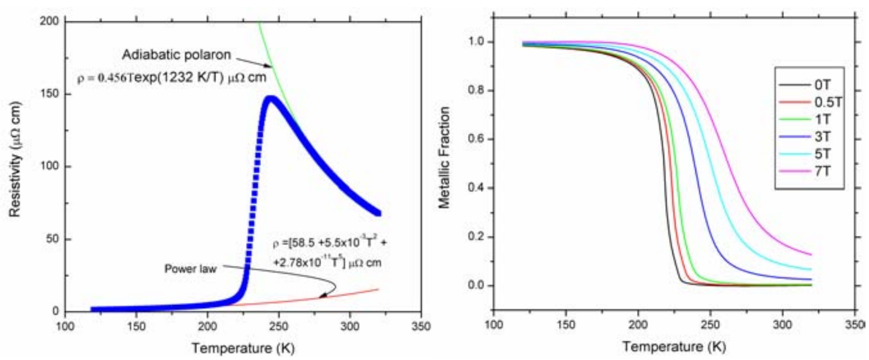
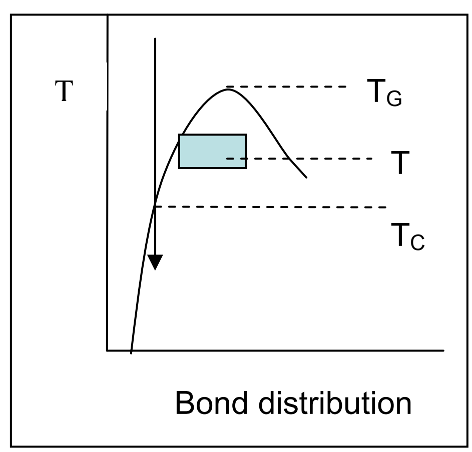


Figure 2. Low temperature and high temperature resistivities and their proportions extracted from a generalized effective medium method

Unlike a conventional percolation picture, the metallic fraction changes significantly with both temperature and field. There is, fortunately, a model that deals with the approach to a percolation-like transition with decreasing temperature, first described by R. Griffiths, and leads to a Griffiths singularity. At any temperature T, some regions are already ordered and form clusters. As the temperature is lowered, the number of clusters grows, but not their size. As a consequence, there is a sudden connection of clusters, leading to percolation—but of a very special sort.

The treatment of the Griffiths phase, that range of temperatures between the maximum possible transition temperature T_G and the observed one at T_C is the focus of the talk. The approach is due to A. J. Bray, and relies on a completely different approach to phase transitions proposed by Lee and Yang (yes, that Lee and Yang). I will show you by a simple example why the partition function for a system undergoing a phase transition can be written as a polynomial with all positive terms, which means that it can be factored into N complex roots, where N is the number of particles in the system. That is, we have $Z = e^{Nh}\Pi_i(z - z_i)$ and further that $F/kT = Nh + \sum_i (z - z_i)$. In both



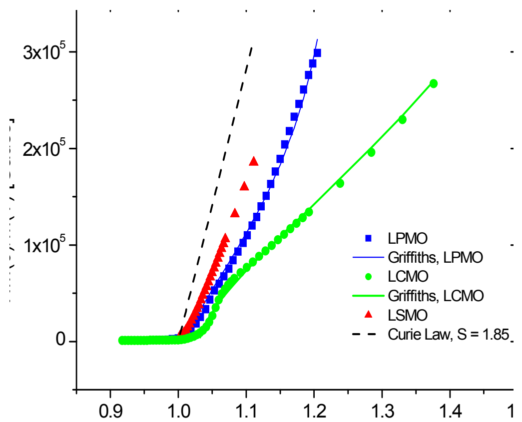
expressions $z = \exp(-2h)$. The remarkable theory of Lee and Yang states that all the z_i lie on the unit circle in the complex plane and can be written as $z_i = \exp(i\theta_i)$, where θ_i measures the angle on the unit circle. Therefore, the problem collapses to determining the distribution of zeroes on the unit circle. If that distribution is described by $g(\theta)$, we can show that the susceptibility is

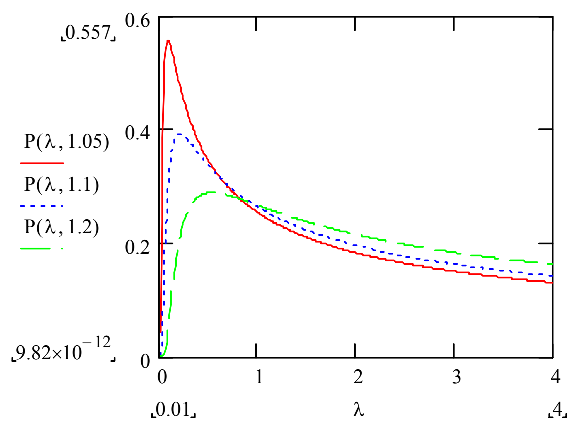
$$\chi = \frac{6C}{T}\int_0^{\pi} \frac{g(\theta,t)}{1-\cos\theta} d\theta.$$

given by

The susceptibility will diverge when there is a finite probability of a zero at $\theta = 0$. The essence of the Griffiths singularity is that there is a pile-up of zeroes near, but not at, $\theta = 0$, but then a sudden collapse of the distribution on to zero.

In the talk, I will discuss a particular model for $g(\theta)$ and show that it reproduces many aspects of the thermodynamics of the CMR materials. In particular, the magnetic susceptibility, the rapid rise of the magnetization at the Curie temperature, and the small entropy associated with the actual transition.





The $g(\theta)$ on the right produces the green susceptibility on the left. For this fit, the Griffiths temperature is 375 K and the actual random transition near 225 K. Further, the lower T_C is, the less entropy change in the vicinity of the transition. The large number of Yang-Lee zeroes at small θ translates into a large number of regions with large susceptibility, but not a single region with a divergent susceptibility. This makes the effective spin large in the Griffiths phase, and releases much of the ordering entropy well above the transition. An interesting direction for high resolution magnetic scattering is to follow the development of these numerous, highly correlated magnetic region and to study their evolution with magnetic field and temperature.