

Magnetic Properties of $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{1-x}\text{Ni}_x\text{O}_3$ Perovskites

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Abstract:

Using the SQUID magnetometer, we studied the magnetic properties of $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{1-x}\text{Ni}_x\text{O}_3$ ($x=0, 0.05, 0.10, 0.20$ and 0.30) perovskites. Both temperature dependent and field dependent magnetic properties show that Curie temperature decreases as a function of doping, ranging from 380K to 300K. Both magnetic measurements and neutron diffraction data hint at long-range magnetic ordering in samples at low temperature and transition phases from paramagnetic to ferromagnetic to antiferromagnetic ordering in samples at room temperature.

1.Introduction

1.1 Paramagnetism

The magnetic moment of an ion is given by

$$\mu = \gamma \hbar J = -g \mu_B J$$

where g is the Landé g -factor, $\mu_B = 9.274 \times 10^{-21}$ erg/Oe is the Bohr magneton, and J is the angular momentum. All atoms have $2J+1$ energy levels. For an electron g is usually approximated to be 2.00, which is the spectroscopic splitting factor. Magnetization is the magnetic dipole moment per volume. Using the methods of statistical mechanics, the paramagnetic magnetization of a solid is given by

$$M = N g J \mu_B B_J(x); (x = g J \mu_B H / kT)$$

where N is the number of ions per unit volume, H is the applied magnetic field, and k is the Boltzmann constant. B_J , the Brillouin function, is defined as

$$B_J(x) = \frac{(2J+1)}{2J} \coth\left(\frac{(2J+1)x}{2J}\right) - \frac{1}{2J} \coth\left(\frac{x}{2J}\right)$$

The equation for M above describes the magnetization of an “ideal” paramagnet. In the classical limit, in which J can have any value, the Brillouin function simplifies to the Langevin function:

$$L(x) = \coth(x) - 1/x$$

If we study the equation for magnetization, we study using Pierre Curie's approximation, which deals with high temperatures and low magnetic fields, that when we increase the temperature and decrease the magnetic field. When we set $x = \mu_B/k_B T \ll 1$, in the Brillouin function, the magnetization reduces to

$$M = N J (J+1) g^2 \mu_B^2 H / 3kT$$

and further to

$$M = C * H / T$$

where C is the Curie constant, defined as

$$C = NJ(J+1)g^2\mu_B^2/3k$$

The magnetic susceptibility is

$$\chi_p = M/H = C/T$$

The lower the temperature, the higher the magnetization gets, and vice-versa.

1.2 Ferromagnetism

Ferromagnetism is the spontaneous magnetization of magnetic moments lining up with each other due to the influence they have on each other. The mean-field approximation derives from the assumption that every ion experiences a field proportional to the magnetization:

$$B_E = \lambda M$$

where λ is a material-specific constant that is independent of temperature. The mean-field approximation is temperature dependent. B_E is assumed to be the equivalent of an exchange field, which is an internal interaction lining up the magnetic moments parallel to each other.

Above the Curie temperature, T_C , the spontaneous magnetization vanishes and the material is paramagnetic. Below the Curie temperature, the material is ferromagnetic. The magnetization is a complicated function of temperature and applied field. We can find T_C by observing λ from the mean-field approximation.

Looking at the paramagnetic phase, we have

$$M = \chi_p (B_A + B_E)$$

where χ_p is the magnetic susceptibility, and B_A is an applied magnetic field which causes a finite magnetization. Assuming that the material is in the paramagnetic phase, we can take Curie's Law for paramagnetic susceptibility to be

$$\chi_p = C/T$$

where C is the Curie constant. If we substitute this into the previous equation, we get

$$MT = C (B_A + \lambda M)$$

and rearranging the variables we can simplify it to

$$\chi = M/B_a = C/(T - C\lambda)$$

We see that the denominator is zero when $T = C\lambda$, in the above equation. At that temperature and below, the magnetization is spontaneous, and because of that we can have a finite magnetization for $B_a = 0$.

From this we get the Curie-Weiss Law

$$\chi = C/(T - T_C) ; \quad T_C = C\lambda$$

which describes the susceptibility variation in the paramagnetic region above the Curie point.

With this information we can define λ to be

$$\lambda = T_C/C$$

1.3 Antiferromagnetism

Antiferromagnetism is like ferromagnetism where instead the magnetic moments line up anti-parallel to each other.

If we begin by assuming separate Curie constants C_A and C_B for ions at two different sites A and B, we will have two interactions as $\mathbf{B}_A = -\mu\mathbf{M}_B$ and $\mathbf{B}_B = -\mu\mathbf{M}_A$; μ is a positive mean field constant. With the mean field approximation we would have

$$M_A T = C_A (B_a - \mu M_A) \quad M_B T = C_B (B_a - \mu M_B)$$

here, the B_a is the applied field. We solve this system of equations using a matrix, by assuming the applied field is zero. The susceptibility to at $T > T_C$

$$\chi = \frac{(M_A + M_B)}{B_a} = \frac{[(C_A + C_B)T - 2\mu C_A C_B]}{(T^2 - T_C^2)}$$

Now, for an antiferromagnetic limit we have $C_A = C_B$. Antiferromagnetism occurs below the

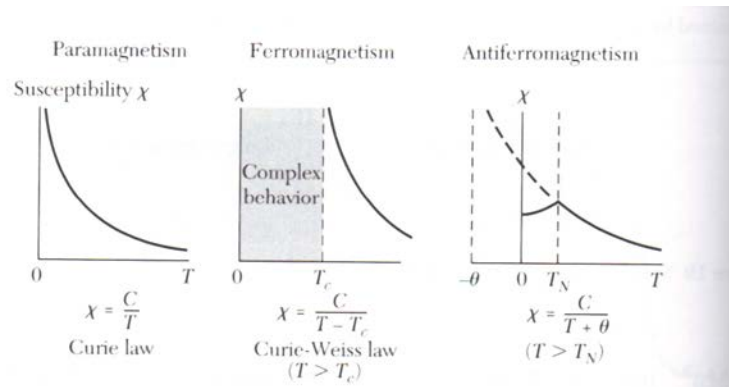
Néel temperature T_N . The Néel temperature is defined by

$$T_N = \mu C$$

after some substitution in the susceptibility equation above, we get

$$\chi = \frac{2C}{(T + \theta)}$$

where θ is $T > T_N$. A summary of all of the magnetizations is shown in the figure below. Note that at T_C for a ferromagnet the magnetization is a singularity, whereas at T_N of an antiferromagnet the magnetization peaks.



2. Methods

For this experiment, we used the SQUID (superconducting quantum interference device), which uses Josephson junctions to measure magnetic fields in samples of $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{1-x}\text{Ni}_x\text{O}_3$. Small samples were obtained from pellets of bulk material of different Ni contents ($x=0, 0.05, 0.1, 0.2, 0.3$). The samples were prepared via solid-state reaction methods. 100 mg bars were cut using a diamond bladed saw. The sample was mounted with cotton into a pill capsule to protect it from moving around. After that, we sewed the capsule into place inside a straw, three-quarters of the way down. We inserted the straw into the SQUID.

3. Results

Magnetization data was obtained using both constant field and variable temperature, as well as varying field at constant temperatures. This data is plotted in two graphs for each sample composition: magnetic susceptibility[χ] vs. Temperature [T], and magnetization[M] vs. applied field[H]. The applied field was in the range of 0-70 KOe. Three sweeps were performed consecutively in the susceptibility measurements: Zero-field cooled (ZFC) where the sample is cooled in 0 field then warmed in a field while taking measurements, field cooled (FC) where the sample is then cooled in the same field while taking measurements, and field cooled warming (FCW) where the field is then turned off and measurements are taken as the sample warms.

As shown in Figures 1, 3, 5, 7 and 9, the transition temperature of the $x=0$ composition is approximately 375K, while for $x=0.05$ it is 330K, for $x=0.1$ it is 350K, for $x=0.2$ it is 300K and for $x=0.3$ it is 300K.

If we look at Figures 2, 4, 6, 8 and 10, we see that there is saturation at the $x=0$ state (at both 5K and 300K), at $x=0.05$ (at 5K, but not at 300K), at $x=0.1$ (at both 5K and 300K), and at $x=0.2$ (at 5K, but not at 300K).

Figure 4 shows a mixture of phases, at the higher temperatures, which can be seen through the combination of saturation and flatness. One point of interest, is that at the $x=0.2$ state, the 300K measurement was the only one we took above the transition temperature. Another point of interest, is at the $x=0.3$ state, there is no saturation at either temperature, even up to 70 KOe. All of the M vs T graphs seem have have comparably similar shapes, however by the time we have 30 percent doping, we see it has a different shape. The properties begin to change and there is a great suppression of the order of magnitude of magnetization, which can be seen on the y-axis, due to doping.

Based off Figure 11, it seems like there is no strong correlation between doping and transition temperature, however it does trend towards T_C suppression as Ni-doping increases.

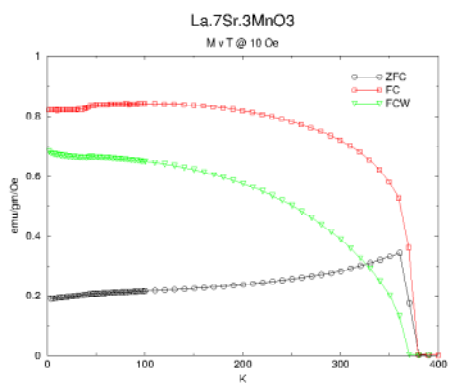


Figure 1: Temperature dependence of susceptibility for $x=0$ measured in a 10 Oe field

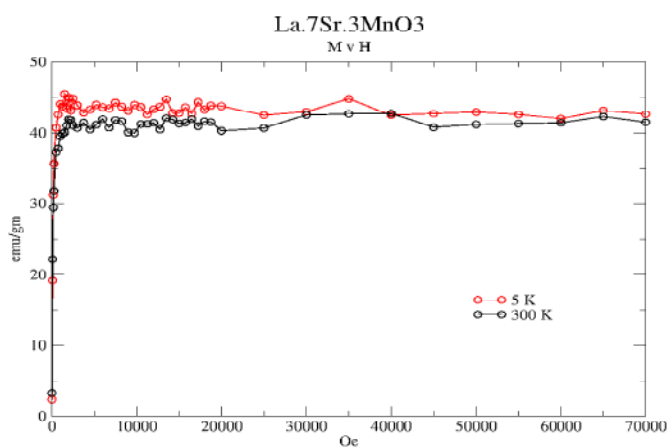


Figure 2: Field dependence of susceptibility for $x=0$ measured in a 10 Oe field

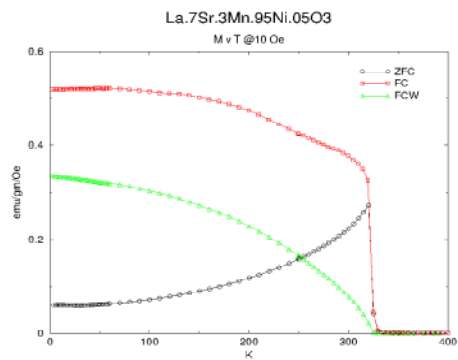


Figure 3: Temperature dependence of susceptibility for $x=0.05$ measured in a 10 Oe field

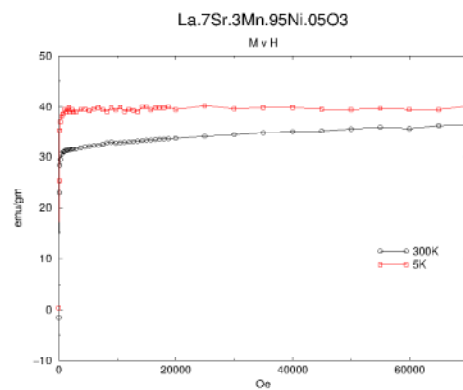


Figure 4: Field dependence of susceptibility for $x=0.05$ measured in a 10 Oe field

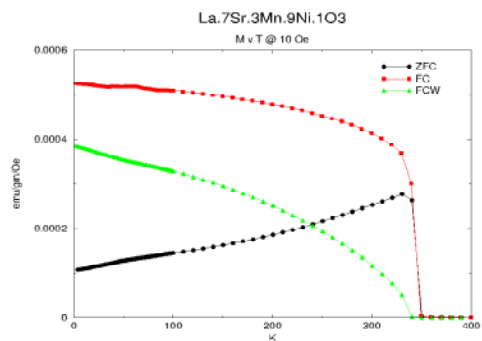


Figure 5: Temperature dependence of susceptibility for $x=0.1$ measured in a 10 Oe field

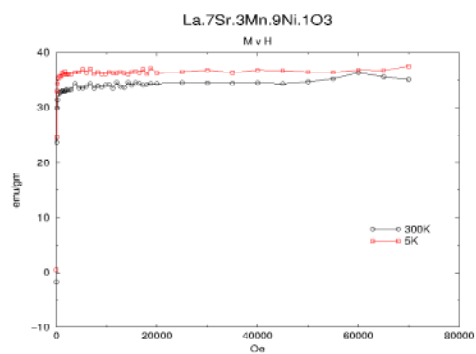


Figure 6: Field dependence of susceptibility for $x=0.1$ measured in 10 Oe field

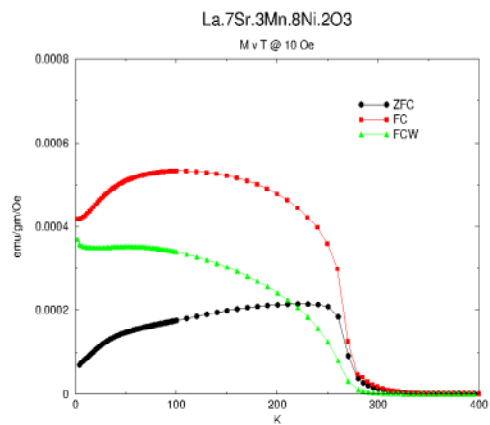


Figure 7: Temperature dependence of susceptibility for $x=0.2$ measured in a 10 Oe field

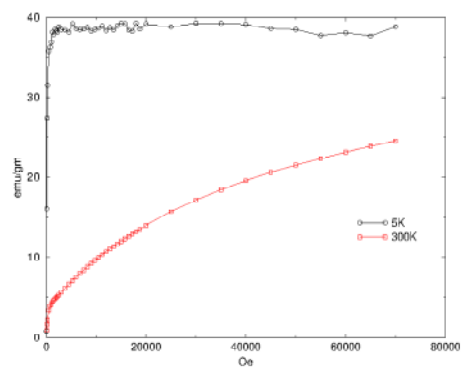


Figure 8: Field dependence of susceptibility for $x=0.2$ measured in a 10 Oe field

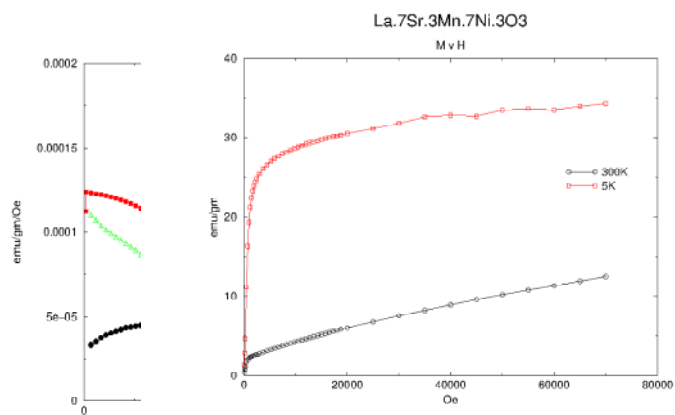
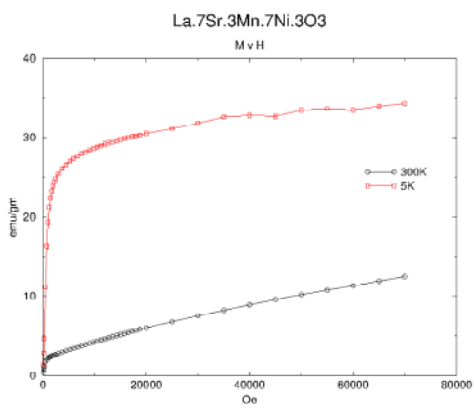


Figure 9: Temperature dependence of susceptibility for $x=0.3$ measured in a 10 Oe field



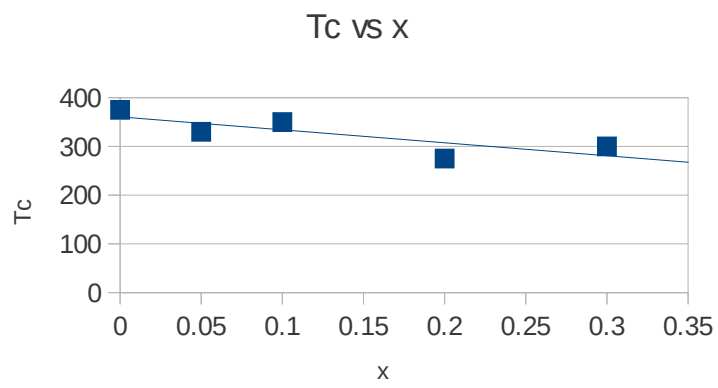


Figure 11: transition temperatures vs x