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Magnetic held dependence of elastic modulus and resistance in $La_{2/3}Ca_{1/3}MnO_3$

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Abstract

We have measured the magnetic field dependence of the anelastic modulus and the electric resistance of a ceramic sample of the magnetoresistant perovskite $La_{2/3}Ca_{1/3}MnO_3$ around the metal-insulator transition by the vibrating reed technique (2.5 KHz). Previous work [1] showed that the modulus becomes higher while the internal friction has a peak at the transition temperature (T_c =262 K). In this work, improvements made on the equipment allowed us to measure at constant deformations (ε <10⁻⁵) and magnetic fields up to 4500 Gauss. We made isothermal measurements of internal friction, modulus and resistance as a function of an applied magnetic field. We found that most of the changes induced by the magnetic field take place in a few degrees (almost 5 K) near the transition temperature where the changes in resistance are more important. Up to the highest magnetic field applied, we found 3% maximum variations of the modulus and no hysteresis while cycling the magnetic field. We suppose that the experiment is placed in the linear response of the inverse Wiedemann effect [2], due to the small deformations used, and that the ferromagnetic domain structure is responsible for the observed effects. Some additional measurements are needed (magnetic hysteresis loops) to be compared with our results. © 2000 Elsevier Science S.A. All rights reserved.

Keywords: Magnetically ordered materials; Chemical synthesis; Scanning microscopy; Elasticity; Magnetostriction

1. Introduction

The colossal magnetoresistant manganese perovskites had been studied for the last years due to the exciting physical properties they show and their possible applications as magnetic sensors. Actually, many authors began to study them in the 50s [3]. The motivation was that the perovskites of the type AMnO₃ are normally, at low temperatures, insulators and develop an antiferromagnetic state. The Mn can exhibit 3+ or 4+ valence depending on the valence of the A ion. In either case, the magnetic interaction between the Mn ions is performed through oxygen, allowing a superexchange interaction to take place. But, if we mix ions of different valence (3+ and 4+) in the place of ion A, we force the Mn ions to have also different valences. In this case another interaction appears, the double exchange, proposed by C. Zener [4] in 1951.

The competition between these two interactions,

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superexchange and double exchange, give rise to many interesting physical properties of these compounds as ferromagnetic and metallic order, colossal magnetoresistance, charge and orbital ordering, etc. The colossal magnetoresistance that they show made them to be studied from the industrial application point of view. Some improvements of C. Zener's theory were made by P.W. Anderson, H. Hasegawa [5] and P.G. de Gennes [6]. Numerical calculations made from microscopic theory and taking into account the exchange interactions failed to explain the transition from metallic to insulator behavior when the compound also develops a ferromagnetic transition. A.J. Millis [7] proposed that a Jahn-Teller deformation of the oxygen octahedron might split the upper level e_a of the 'd' electrons of Mn, increasing the localization of the electrons and lowering the resistivity when we increase the temperature. This kind of deformations of the lattice and the magnetostriction (due to the magnetic order that take place at the metal-insulator transition) make these compounds very interesting due to their anelastic properties. Then, we studied the magnetoresistant perovskite La_{2/3}Ca_{1/3}MnO₃ around the magnetic transition, measuring the resistance, internal friction and anelastic modulus as a function of temperature and the applied magnetic field.

2. Experimental details

Measurements were done in a conventional vibrating reed apparatus capable of keeping constant the measuring amplitude of deformation, changing the temperature of the sample between 40 and 300 K and applying, from a superconducting coil, a magnetic field up to 4500 Gauss. The vibrating reed apparatus is made of one driving electrode and one detecting electrode at one end of the sample. The other end of the sample is fixed with silver paint to a bulk copper piece. Three thin gold wires (25 μm) were attached at one side of the sample to to measure the resistance through a four-terminal configuration. Fig. 1 shows a schema of the assembly of the sample $(0.1 \times 1 \times 6$ mm³). Sample free length of vibration was 6 mm and its oscillating frequency around 2500 Hz. According to this technique the anelastic modulus M and the internal friction IF are calculated from the oscillating frequency (f), the driving voltage $(V_{\text{driv.}})$ and the amplitude of oscillation (b)

$$M \sim f^2$$
 $IF \sim (V_{\text{driv}}/f)^2 1/b$

It is not possible to measure the absolute value of the deformation induced by the driving electrode, but it was estimated numerically that the deformation was below 10^{-5} . The ceramic sample was prepared in the convention-

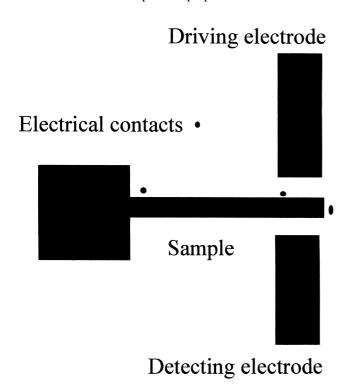


Fig. 1. Experimental set-up.

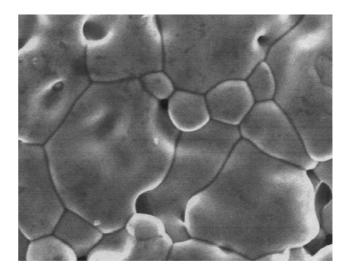


Fig. 2. SEM image of $La_{2/3}Ca_{1/3}MnO_3$ (photograph width=60 µm).

al solid state solution. Successive annealing at 1300°C during 60, 50 and 100 h were performed, and after each cycle the sample was ground and pressed at 5 Tn. Finally it was placed during 24 h in an oxygen atmosphere at 1200°C . The finished sample has very large grains (5–25 μ m) as it shows Fig. 2. There are no many holes or empty regions and the grains have a large surface of contact between them. This point made us think that we could measure microscopic properties rather than macroscopic ones.

3. Results and discussion

Isothermal measurements of the electrical resistance, the internal friction and the elastic modulus as a function of the applied magnetic field are shown in Figs. 3–5. Fig. 3 shows the metal–insulator transition at $T_{\rm c}$ and the high magnetoresistance of the sample upon applying the mag-

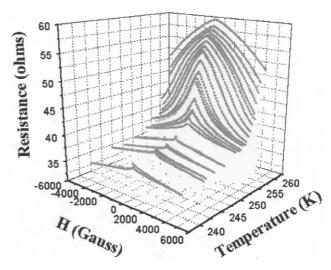


Fig. 3. Electrical resistance vs. magnetic field and temperature.

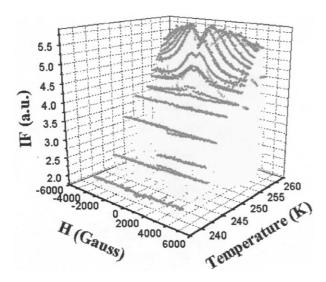


Fig. 4. Internal friction vs. magnetic field and temperature.

netic field. In [1] we have shown the IF peak and the modulus step (vs. temperature) developed at $T_{\rm c}$. In Fig. 4 we can see only the increasing part of this IF peak taking into account the isotherm values at H=0 Gauss. The decreasing part is masked by the lower temperature data. Furthermore, in Fig. 4 we see that, in addition to the usual internal friction peak obtained at $T_{\rm c}$ with no applied field, there is another IF peak centered at different magnetic fields which moves with temperature. At the 255 K isotherm this IF peak is centered at 800 G. It moves to 3200 G at the 257.5 K isotherm. This important shift of the IF maximum with magnetic field in such a narrow temperature range, shows the high sensitivity of the magneto striction to the temperature as well as to the magnetic field. Corresponding to this peak, the modulus shows changes in its slope versus the magnetic field, as illustrated in Fig. 5. At the 245 K and 260 K isotherms we

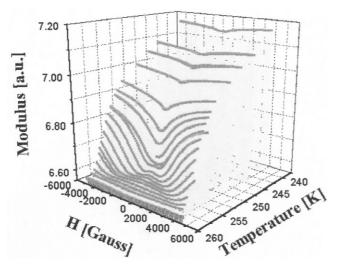


Fig. 5. Elastic modulus vs. magnetic field and temperature.

observed no *IF* peak as a function of the magnetic field. We also see, from Fig. 5, that the modulus increases below the transition temperature, as it does the magnetization when we apply a magnetic field.

These facts indicate that some process is taking place versus the applied field at the transition temperature. When the temperature decreases below $T_{\rm c}$, a negative magneto striction takes place [8]. We suppose that this fact is related to the increasing value of the modulus. This is supported by the enhancement of the modulus when we increase the applied magnetic field and induce an additional magneto striction. This change is also high at the temperature where the magnetoresistance has its highest values, indicating a close relationship between electronic, magnetic and elastic properties.

S. Chikazumi [2] page 437, has shown that it is not possible to obtain a higher modulus due to a volume magneto striction, actually the modulus decreases when a volumetric magneto striction happens. That means, as the microscopic origin of the volumetric magnetostriction is mainly due to the exchange interaction and higher order terms of the magnetic anisotropy, these interactions cannot be responsible for the increase of the modulus at $T_{\rm c}$. Only low order terms of the magnetic anisotropy might increase the modulus at $T_{\rm c}$.

We have not, at present, a clear idea of the origin of the IF peak which appears near $T_{\rm c}$ and moves with magnetic field. But, probably it has the same origin that the internal friction peak which we find at $T_{\rm c}$ as a function of temperature. The magnetic field induces ferromagnetism at temperatures above $T_{\rm c}$ and due to this magnetization, it appears a magnetostriction deformation which is detected through the elastic properties.

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