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# Anomalous magnetic field dependence of $T_{AF}$ in $La_{0.95}Sr_{0.05}MnO_3$

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**Abstract.** The magneto-elastic properties of single-crystalline $La_{0.95}Sr_{0.05}MnO_3$ have been studied ultrasonically. Our investigations focussed on the temperature interval where magnetic ordering starts to evolve and results in a spin canted antiferromagnetic ground state. In detail the experiments revealed that the magnetic order parameter in low-doped manganite is only weakly coupled to lattice strains. Furthermore, the anomalous temperature dependence of the order parameter as found resembles highly that in stoichiometric $LaMnO_3$. However, the main and most surprising finding is that external magnetic fields favor the spin canted phase in $La_{0.95}Sr_{0.05}MnO_3$. It is unclear at present how the exchange interaction can be tuned by magnetic fields in the way observed and we are not aware of existing theoretical concepts which might give a plausible explanation for the unexpected field dependent behavior of the critical temperature. We believe, however, that this behavior primarily results from the fact that the exchange interaction depends sensitively on the orbital configuration of the manganese $d$ electrons.

**PACS.** 75.50.Ee Antiferromagnetics – 62.65.+k Acoustical properties of solids – 75.40.Cx Static properties (order parameter, static susceptibility, heat capacities, critical exponents, etc.)

## 1 Introduction

Since a couple of years manganites with perovskite related crystal structure attract much attention due to the rich variety of ordering phenomena found in these materials giving rise to peculiar magnetic and unusual transport properties (for a comprehensive overview see *e.g.* [1]). As became clear early the properties depend sensitively on the charge imbalance between the mean cation valency and the doubly ionized oxygen – the valency ratio is easily adjusted by substitutional cation doping. It is common belief that the complex phase diagram of manganites originates from the competition between carrier delocalization in the $Mn^{+3}/Mn^{+4}$-sublattice mediated by the double-exchange mechanism [2–4] and local charge/orbital ordering supported or driven by Coulomb interactions or the Jahn-Teller mechanism [5–8]. Depending on which interaction is dominating either an antiferromagnetic insulator or a ferromagnetic metal is expected to be the ground state. However, as past experience has shown [9–14] the behavior of manganites is of much higher complexity than originally assumed [15,16] and to get a clear idea of the underlying physics turns out to be a difficult task in the mean time.

Concerning the particular case of Sr-doped $LaMnO_3$, a lot of experimental data are already available. Nevertheless, a fundamental understanding of the unusual physical properties met in this compound is still lacking as well. From the theoretical point of view, there is still disagreement about the relative importance of electron-lattice and Coulomb interactions as well as magnetic and electron-phonon effects. From the experimental side, however, there is also some lack of clarity about the role of the electron-lattice interaction and its dependence on the Sr concentration, in particular. Generally, the coupling between electrons and lattice is certainly reflected in the elastic properties which, however, can be investigated quite well ultrasonically. Very surprising, only poor and sketchy knowledge about the elastic behavior of Sr-doped manganite is available from the current literature [17,18]. Quite a few published results refer to investigations performed particularly on single crystals [19,20] and to our knowledge no magnetic field dependent measurements have been reported so far.

The physical properties of doped manganite depend strongly on chemical composition and their field and temperature dependent behavior reflect the complex interplay of the various involved interactions in a subtle manner. Taking this difficulty into account we confine here ourselves to investigations of the low-doped compound $La_{0.95}Sr_{0.05}MnO_3$. For this dopant concentration the interactions caused by $Mn^{+3}/Mn^{+4}$ valency mixing are just starting to evolve but are still in its infancy state. In order to shed some light on the role of the electron-lattice interaction in this doping regime temperature and field dependent sound velocity measurements were done. In particular, we studied the magneto-elastic

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interaction in the vicinity of the antiferromagnetic transition in $La_{0.95}Sr_{0.05}MnO_3$.

## 2 Experimental

The ultrasonic and magnetic investigations were performed on a $La_{0.95}Sr_{0.05}MnO_3$ single crystal some mm in length which was grown by the floating zone method with radiation heating under Ar atmosphere. The orientation of the nearly twin-free crystal was determined by Laue X-ray diffraction. After cutting, a pair of parallel end faces was polished to optical flatness. Orientation dependent measurements of the magnetization were done by means of a SQUID magnetometer. Longitudinal and transverse ultrasonic waves were generated and received using overtone polished X- and AC-cut quartz transducers which were carefully bonded to one of the sample end faces with a polysulfide liquid polymer. A high resolution sampled-continuous wave spectrometer was used to perform temperature and field dependent attenuation and sound velocity measurements at frequencies between 10 and 30 MHz. The samples were cooled down from ambient to liquid He temperatures and warmed up again quite slowly with a rate of less than 1 K/min in order to avoid crystal damaging due to thermal stress and structural changes being possibly associated with phase transitions taking place.

## 3 Results

### 3.1 Doping enhanced spin canting

At ambient temperatures Sr-doped lanthanum manganite $La_{1-x}Sr_xMnO_3$ adopts the crystal structure of an orthorhombically distorted perovskite for a Sr content $< 0.1$/mole. The orthorhombicity is largest for stoichiometric $LaMnO_3$ and decreases monotonously on doping which strongly affects the Jahn-Teller driven lattice distortion. The latter fact also accounts for the steady decrease of the critical temperature of the structural transition to the orthorhombic phase above room temperature. The concomitant decrease of the magnetic transition temperature on doping is quite moderate in comparison with that of the structural transition [21–24]. In addition, however, doping also modifies the magnetic structure and favors ferromagnetic alignment of the ordering moments [25].

Neutron diffraction studies on pure LaMnO3 [26,27] revealed ferromagnetic ordering in the orthorhombic $a$-$b$ plane but antiferromagnetic coupling of successive planes along the $c$-axis (Pbnm space group) below 139.5 K. Though slightly canted the ordered moments are almost parallel to the $b$-axis. The magnetization in $c$-direction due to spin canting may therefore be considered as a secondary order parameter. Remarkably, the canting angle depends sensitively on external magnetic fields giving rise to a considerable magnetic susceptibility also in the ordered state.

The magnetization of single-crystalline $La_{0.95}Sr_{0.05}MnO_3$ accompanying the evolution of the antiferromagnetic phase is presented in Figure 1. From these data

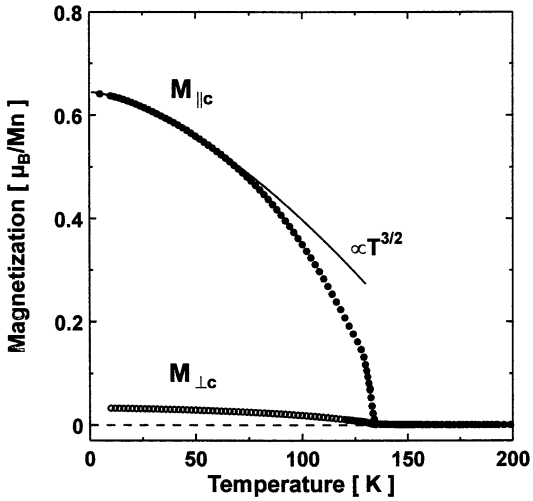


**Fig. 1.** Magnetization of single-crystalline $La_{0.95}Sr_{0.05}MnO_3$ measured parallel and perpendicular to the orthorhombic $c$-axis in a magnetic field of 10 mT. The low-temperature behavior deduced from spin wave theory is indicated by a solid line.

a critical temperature $T_{AF} = 133.5 \pm 0.5$ K is obtained. At a first glance, the overall temperature dependence of the magnetization is very reminiscent of stoichiometric $LaMnO_3$. The "weak" ferromagnetic moment of 0.64 $\mu_B$/Mn in $c$-direction achieved at low temperatures, however, is about four times larger than the respective value in the undoped compound [28]. Since the local moments are only slightly altered on doping the increase of ferromagnetism should mainly result from stronger spin canting. Thus, assuming a mean manganese moment of about 3.8 $\mu_B$ [25,26] in $La_{0.95}Sr_{0.05}MnO_3$, a canting angle of roughly 10° can be estimated from our data.

The existence of propagating spin waves in $LaMnO_3$ was fully confirmed by inelastic neutron scattering [26]. Their presence usually gives rise to a moderate increase of the temperature dependent magnetization also far below the critical temperature. Remarkably, a magnetization behavior $\propto T^{3/2}$ reminiscent of ferromagnetic spin waves is observed in the Sr-doped crystal as emphasized in Figure 1.

In addition, the presented data reveal that the magnetic anisotropy of our sample is almost perfect. Below $T_{AF}$ the magnetization nearly vanishes for directions perpendicular to the orthorhombic $c$-axis. A significant reduction of the magnetic anisotropy might arise from structural domains caused by [110]-[001] twinning of the crystal (called $F$ domains in [26]). Therefore a strong magnetic anisotropy indicates that only small amounts of such defects if ever are present in our sample and confirms the high crystal quality as already established by Laue X-ray diffraction.

### 3.2 Magneto-elastic properties of the spin canted phase

The study of the propagation of ultrasonic waves in solids when passing through the critical temperature into an ordered state has proven to be a valuable tool for investigating the coupling between strain and order parameter as well as the critical dynamics in the vicinity of the transition temperature [29,30]. Attenuation and sound velocity measurements of both longitudinal and transverse sound waves were performed in order to get an idea of the strength of the magneto-elastic coupling attributed to the antiferromagnetic ordering in $La_{0.95}Sr_{0.05}MnO_3$.

In detail we studied the elastic behavior by sound waves which propagate along the orthorhombic $b$-axis, *i.e.* almost in direction of the ordered magnetic moments. Both the longitudinal $C_{22}$- and the transverse $C_{55}$ -mode show small but distinct anomalies right at the transition temperature. After substraction of the regular temperature dependent background as sketched in the inset of Figure 2, we obtain the elastic contribution connected with the magnetic ordering also shown there enlarged. For the ordered state both sound modes indicate a pronounced stiffening of quite similar magnitude. A steplike feature at $T_{AF}$ as it is observed for longitudinal sound, however, is absent in the temperature dependence of the transverse mode. Qualitatively, elastic anomalies of the type shown in Figure 2 are familiar from many cases in solid state physics. They suggest a phase transition which is most probably of second order and which is not primarily caused by symmetry breaking of the lattice unit cell. The observed transition behavior is often found also in magnetic systems and reminiscent of antiferromagnetic transitions like in $MnF_2$ [31], for instance.

Simultaneous measurements of the temperature dependent ultrasonic attenuation (not shown here) revealed that sound dissipation due to critical fluctuations does not play any significant role in the vicinity of $T_{AF}$. Thus mean field theory is justified and should be fully sufficient in order to give a quantitative treatment of the elastic behavior. Careful comparison of the sound data obtained from slow cooling and heating runs, however, indicated slightly hysteretic behavior (0.2–0.3 K) suggesting a weak first order of the antiferromagnetic transition in $La_{0.95}Sr_{0.05}MnO_3$.

In the case of antiferromagnetic ordering there exists due to symmetry reasons no linear coupling between the elastic strain and the order parameter which is the sublattice magnetization $M_{AF}$. Therefore the leading interaction term in the free energy density depends at least quadratically on $M_{AF}$. On the other hand, it is generally sufficient to restrict oneself to an interaction energy of the form

$$f_{\text{int}} = \sum_{\Gamma} \left\{ h_\Gamma M_{AF}^2 \varepsilon_\Gamma + k_\Gamma M_{AF}^2 \varepsilon_\Gamma^2 \right\} \qquad (1)$$

where $h_\Gamma$ and $k_\Gamma$ denote coupling coefficients belonging to a strain of symmetry $\Gamma$. Since fluctuations are not relevant in our case, there should be no effect on the elasticity above $T_{AF}$. A non-zero elastic contribution is expected in mean field approximation from equation (1) only for the ordered phase *i.e.* below the critical temperature. For the low-frequency limit the calculation yields

$$\Delta C_\Gamma = -2h_\Gamma^2/b + 2k_\Gamma \langle M_{AF} \rangle^2 \text{ for } T \leq T_{AF} \qquad (2)$$

where $b$ comes from the quartic term in the Landau free energy expansion of the order parameter. It turns out that equation (2) is appropriate to describe the experimental findings of $La_{0.95}Sr_{0.05}MnO_3$ satisfactorily.

Since the mean value of $M_{AF}$ vanishes for $T \geq T_{AF}$ no contribution is expected from the second term in equation (2) right at the transition temperature. Due to the first term, however, a sudden change may occur in the temperature dependent stiffness coefficients giving rise to a steplike feature at $T_{AF}$. It is this behavior what is

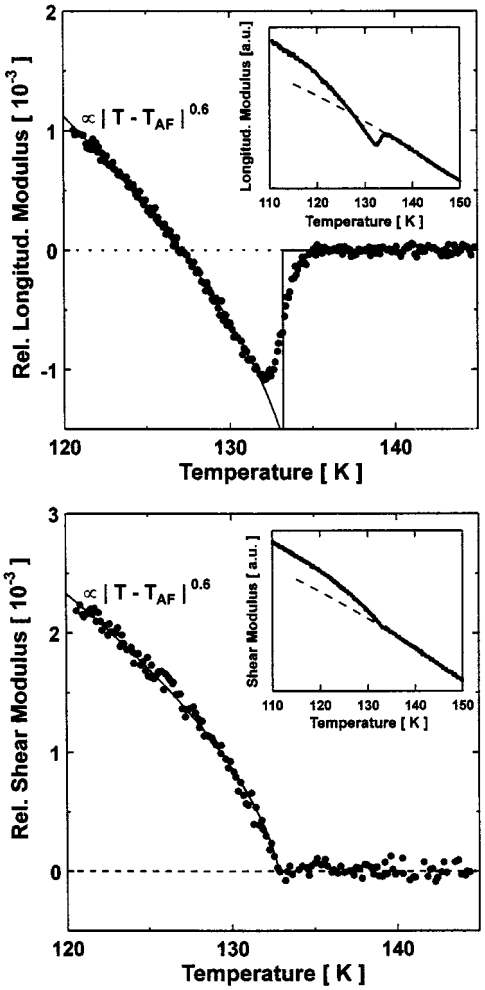


**Fig. 2.** Temperature dependence of the normalized elastic moduli in the vicinity of the antiferromagnetic transition in $La_{0.95}Sr_{0.05}MnO_3$ obtained for the longitudinal $C_{22}$-mode (upper panel) and transverse $C_{55}$-mode (lower panel) after substracting the elastic background as shown in the insets. The step anomaly for longitudinal strain theoretically expected in the static limit is marked by solid lines. For both modes the temperature dependence is well approximated by the power law $|T - T_{AF}|^{0.6}$ below the critical temperature.

actually observed for the longitudinal but not for the transverse mode. This gives some evidence for the assumption that the observed jump in stiffness is primarily caused by volume coupling $\propto h_{\Gamma 1}$ whereas the shear coupling parameters appear to be indiscernible within experimental resolution.

In contrast both longitudinal and transverse sound couple equally to the evolving order parameter for $T < T_{\rm AF}$. From the elastic data shown in Figure 2 it turns out that the coupling parameters $k_\Gamma$ are of nearly the same order. Furthermore, as indicated by continuous lines the temperature dependence for $T < T_{\rm AF}$ is nicely approximated by a power law which is the same for both modes as well. According to equation (2), the elastic response is proportional to the square of the order parameter. Thus its temperature dependent behavior can be easily determined from our sound data. For $La_{0.95}Sr_{0.05}MnO_3$ we got this way

$$\langle M_{\rm AF}\rangle \propto |T/T_{\rm AF} - 1|^\beta,\ \beta = 0.30 \pm 0.02. \qquad (3)$$

The critical exponent we obtained differs significantly from that theoretically expected for an isotropic 3-dimensional Heisenberg model where $\beta = 0.36$. Within the margin of error, however, our result fairly agrees with the exponents determined by means of neutron scattering not only in pure $LaMnO_3$ ($\beta = 0.28 \pm 0.01$ [26]) but also in the ferromagnetic compound $La_{0.7}Sr_{0.3}MnO_3$ ($\beta = 0.295 \pm 0.002$ [32]). It is commonly believed that the low value of $\beta$ arises from a strong single ion anisotropy and can be convincingly explained, provided that the anisotropy is properly taken into account from theory.

### 3.3 Field tuned exchange interaction

It is well-known from investigations over a broad range of dopant concentrations that the magnetic and the transport properties of Sr-doped manganites are closely connected with the lattice degrees of freedom including charge and orbital ordering. In addition, however, it also became clear that magnetic fields play a significant role in so far as the subtle interplay between spin, charge and orbital degrees of freedom can be easily modified by them. For instance, it has been found that the structural transition to the orthorhombic O' phase driven by the cooperative Jahn-Teller effect at higher temperatures as well as the transition to a charge ordered state taking place at lower temperatures are markedly influenced by externally applied magnetic fields. Whereas the structural transition is suppressed by about 1 K/T [12], charge ordering, in contrast, is supported by external fields and the transition temperature increases considerably ($\approx$ 2...4 K/T, see *e.g.* [12,33]). When cooling down to liquid He temperatures we didn't find any indications for an additional phase transition which might have been attributed to charge ordering in our low-doped sample. This finding, however, is in full agreement with results published previously [22,23,34]. There, it has been established that charge ordering in Sr-doped manganite does not occur for dopant concentrations less than 8–10% Sr/mole.

It is not clear at present whether there is any special influence of external fields also hidden in the magnetic phase diagram of low-doped manganite. Although numerous field dependent investigations of the antiferromagnetic state in pure and lightly doped samples were recently performed [35,36], they solely focussed on tracking down the spin-flop transition expected at higher magnetic field strength of more than 10 T. To our knowledge, no information concerning the influence of external fields on the antiferromagnetic transition is available from the current literature so far. Experimentally an important drawback of magnetization measurements is that the resolution power at higher field strength suffers considerably from the strongly increasing susceptibility on approaching the critical temperature. The elastic coefficients, however, show rather sharp anomalies right at the transition temperature as shown before. Hence, field dependent investigations of the antiferromagnetic transition can be accurately performed by means of ultrasound.

A representative collection of data sets obtained from longitudinal sound velocity measurements at various field strengths is presented at the top of Figure 3. The magnetic field points here in the direction of the orthorhombic *c*-axis. As is clearly seen, the elastic anomaly associated with the onset of antiferromagnetic ordering markedly changes when an external field is applied. Most remarkable, with increasing field strength the anomaly unambiguously shifts towards higher temperatures. In order to describe this puzzling behavior more accurately we choose the midpoint of the steplike change in the elastic modulus to define a proper transition temperature. The field dependence of $T_{\rm AF}$ shown at the bottom of Figure 3 is obtained this way. For field orientation $\mathbf{B} \parallel \mathbf{c}$ a positive coefficient of about 2 K/T is determined from these data by linear approximation. Quite in contrast to this case, it was impossible, however, to detect even slightest changes in the overall elastic behavior for perpendicular field orientations. This finding is also indicated in Figure 3. For the sake of completeness it should be added that analogous investigations performed separately by using transverse sound waves fully confirm these results.

In view of the data presented in Figure 3 it can be fairly assumed that the dependence of the exchange interaction between the manganese moments on the field magnitude is linear in lowest order for $\mathbf{B} \parallel \mathbf{c}$. Since $T_{\rm AF}$ remains unchanged for $\mathbf{B} \perp \mathbf{c}$, however, the exchange coupling seems to be preferentially influenced by the field component directed along the antiferromagnetic wave vector running parallel to the crystalline *c*-axis. Hence, it seems further likely to assume that the relevant contribution is given by $|\mathbf{B}|\cos\theta - \theta$ denotes the polar angle with reference to the *c*-axis. This expectation is indeed fully met by experiment as becomes evident from the angular dependence shown in Figure 4.

Summing up, the antiferromagnetic transition in $La_{0.95}Sr_{0.05}MnO_3$ depends significantly on both magnitude and orientation of externally applied magnetic fields. Needless to say, the pronounced field dependent increase of $T_{\rm AF}$ is very surprising, because it would never be

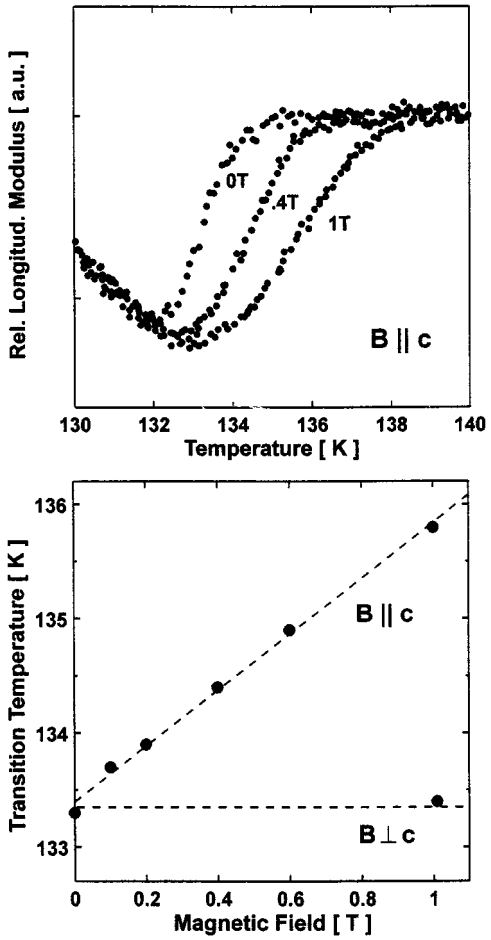


**Fig. 3.** Upper panel: Elastic anomaly attributed to the antiferromagnetic transition in $La_{0.95}Sr_{0.05}MnO_3$ for varying magnetic fields along the crystalline *c*-direction. Lower panel: Field dependence of the transition temperature for field directions parallel and perpendicular to the *c*-axis – dashed drawn lines are guides to the eye.

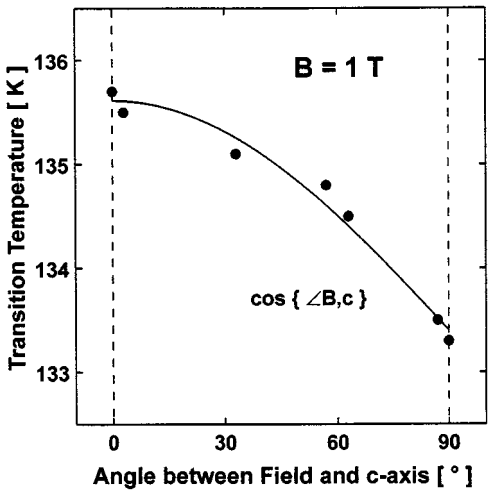


**Fig. 4.** Transition temperature at a constant magnetic field strength of 1 T depending on the field orientation in reference to the crystalline *c*-axis.

expected for a conventional antiferromagnet. It is this particular finding which has to be considered from any theory proposed for low-doped manganites. However, to find an appropriate description for this peculiar behavior based on the exchange mechanisms involved in these compounds seems to be a bigger challenge.

From the theoretical point of view, various contributions to the exchange interaction are expected in doped manganite. Apart from the familiar symmetric Heisenberg Hamiltonian there are two anisotropy terms which have to be taken into account: one arises from the spin-orbit coupling under the local crystalline electric field at the manganese sites, the other comes from the antisymmetric exchange introduced first by Dzyaloshinskii and Moriya [37,38]. Both terms are estimated to be about one order smaller than the symmetric exchange term [28] and should be therefore of minor importance. On the other hand, the evaluation of the spin-orbit term mostly refers to a *d* level splitting obtained for cubic ligand fields. Since the octahedral environment of the manganese ions is strongly distorted and as been proven of orthorhombic symmetry [39,40], the assumed insignificance of spin-orbit coupling, however, seems to be at least questionable. Furthermore, mobile $Mn^{4+}$-holes can give rise to double-exchange interactions favoring ferromagnetic coupling along the antiferromagnetic chain axis. This interaction term depends on both the exchange and transfer integral as was explicitly shown by de Gennes [4].

Due to symmetry reasons one might speculate that the pronounced field dependent anisotropy is intimately related to perturbations of the non-degenerate orbital 3*d* states. Provided that the orbital configuration is changed by an external field preferentially for certain field directions, each of the involved exchange mechanisms should be more or less influenced, though it can be hardly settled at first glance which of them actually dominates the ordering process. According to this model the anomalous field dependence of $T_{AF}$ would reveal a delicate balance between competing coupling terms associated with superexchange and probably double-exchange interaction which, however, depends sensitively on the respective orbital configuration.

On the other hand, it might be questioned whether it is justified in our case to assume a homogeneous system. There are hints in the literature that trapping of $Mn^{4+}$-holes may indeed play a role in lightly doped manganites [41–43]. Theoretically, the existence of bound states giving rise to local inhomogeneities in the spin polarization has been already discussed by de Gennes [4]. More recently, the formation of an inhomogeneous state due to electronic phase separation has been predicted for doped manganites [44–47] and it is a common belief that particularly the canted phase of the doped compound is unstable against segregation of small ferromagnetic "droplets" embedded in an antiferromagnetic host. Ultrasound probes bulk properties and it is beyond doubt that the elastic anomalies accompanying the transition in $La_{0.95}Sr_{0.05}MnO_3$ reflect bulk behavior. Clear indications of coexisting phases, however, are missing. For instance, no

splitting into two or more transitions was found. Therefore it seems likely that phase separation in this compound is less important which should be most probably attributed to the low dopant concentration. Nevertheless, a situation as described by de Gennes is fully within the bounds of possibility and can be hardly ruled out by our experimental findings.

## 4 Conclusions

Ultrasonic experiments performed on single crystalline $La_{0.95}Sr_{0.05}MnO_3$ clearly show that the transitional behavior to a canted antiferromagnetic ground state is only marginally influenced by the electron-lattice interaction in low-doped manganites. The elastic behavior around the transition temperature $T_{AF}$ indicates a weakly strain coupled order parameter. Indeed, both magnitude and size of the observed anomalies are reminiscent of phase transitions found in conventional antiferromagnetic insulators, for instance. This experimental finding is somehow surprising because strong magneto-elastic effects may be expected already in low-doped compounds due to the intimate relationship between lattice and magnetic properties as suggested from their pronounced dependence on substitutional doping. However, there are hints from field dependent investigations which point to a more subtle interplay between lattice and electronic degrees of freedom. The most astonishing result to be mentioned is the fact that external magnetic fields favor antiferromagnetic ordering in $La_{0.95}Sr_{0.05}MnO_3$. Apart from a strong field dependent anisotropy it becomes obvious from our investigations that magnetic fields directed along the crystalline $c$-axis not only enforces stronger ferromagnetic alignment of the antiferromagnetically ordered moments but also lead to a lower ground state energy.

In our opinion it seems to be a difficult task to find an appropriate model which lucidly explains on a microscopic level the quite unexpected properties of the antiferromagnetic ordering in this system. We believe, however, that the peculiar field dependent behavior most probably originates in a sensitive dependence of the exchange interaction on the orbital configuration of the manganese $d$ electrons. Though the local orbitals should be modified by external fields only weakly, simultaneous changes of the exchange coupling between neighboring manganese ions might be nevertheless strong because it depends sensitively on the wavefunctions overlap. Surely, a deeper theoretical understanding of the underlying physics suffers from the complexity of the interactions which have to be considered properly and is, of course, far beyond the scope of our report.

In this context, however, special attention should be furthermore paid to the fact that the pronounced field dependent increase of $T_{AF}$ is very reminiscent of a quite similar phenomenon found in heavier doped samples for the transition to a charge ordered state. Indeed, in both cases the field dependent shift of the transition temperature is positive and of the same order. Therefore it can be fairly assumed that the mechanism responsible for the peculiar transitional behavior found in doped compounds with a Sr content $\geq 0.1$ is already existing in $La_{0.95}Sr_{0.05}MnO_3$.

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