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Orbital ordering in cubic LaMnO₃ from first principles calculations

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Abstract. We report on first principles self-interaction corrected LSD (SIC-LSD) calculations of electronic structure of LaMnO₃ in the cubic phase. We found a strong tendency to localization of the Mn e_g electron and to orbital ordering. We found the ground state to be orbitally ordered with a staggered order of $x^2 - z^2$ and $y^2 - z^2$ orbits in one plane and this order is repeated along the third direction. The difference in energy with a solution consisting of the ordering of $3x^2 - r^2$ and $3y^2 - r^2$ is, however, very small. The system is in the insulating A-type antiferromagnetic ordered state in both cases. The presence of orbital ordering means breaking of the cubic symmetry and without recourse to distortion. The latter may rather be the result of the orbital ordering but the symmetry of this ordering is determined by coupling to the lattice. The strong tendency to localization of the e_g electron in LaMnO₃ accounts for the survival of local distortions above the structural phase transition temperature.

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1. Introduction

Orbital ordering in LaMnO_3 has always been associated with the Jahn–Teller instability of the system as a result of the degeneracy of the e_g orbitals. Because of the crystal field due to the oxygen octahedron the t_{2g} orbitals are known, from a local picture, to lie lower in energy than the e_g ones. The t_{2g} orbitals are occupied each by one electron and because of strong intraatomic Hund exchange interaction the spins of these electrons are aligned parallel forming a $S = 3/2$ spin which is sometimes treated as a classical spin in model calculations. In LaMnO_3 there is one more electron that occupies an e_g orbital. Because the two e_g orbitals are degenerate, the system is unstable towards a distortion which would lift the degeneracy. The amount by which the system is distorted is then determined by the competition between the gain in the electronic energy and the increase in the elastic energy of the lattice due to distortion. LaMnO_3 is found in the distorted phase below 780 K. Orbital ordering in LaMnO_3 has been observed by resonant x-ray scattering on the Mn-K edge. It was found that this ordering decreases above the Néel temperature 140 K and disappears above $T = 780$ K concomitant with a structural phase transition [1].

In 3d transition metal compounds with orbital degeneracy two scenarios are invoked to explain orbital ordering. On the one hand there is superexchange interaction between orbitals on different sites. This involves virtual transfer of electrons and strong on-site electron–electron interaction. On the other hand, cooperative JT distortions or electron–lattice interaction leads to splitting of the degenerate orbitals and thus to orbital ordering. Although the ground-state ordering of LaMnO_3 can be explained by both mechanisms it is not easy to say which is the dominant contribution. This question may sound of little importance so far as LaMnO_3 is concerned but it is important to know the answer because whichever is the dominating mechanism will remain more or less active once the system is doped. In which case the two mechanisms, i.e., electron–lattice interactions or electron–electron correlations may lead to different physics for the doped systems.

There have been attempts, by using model calculation, at explaining how orbital ordering can occur if one assumed an antiferromagnetic spin ordering [2]. However, as mentioned above, temperatures at which the orbital ordering sets in are much higher than the Néel temperature of A-AF spin ordering. Orbital ordering cannot, therefore, be attributed to spin ordering. In previous LSD [3]–[5] and model HF [6, 7] calculations it is found that inclusion of distortions is necessary to recover the correct A-AF and insulating character of LaMnO_3 in the ground state. The cubic system was found to be both metallic and ferromagnetic in the LSD calculations. By using self-interaction correction (SIC) to the LSD we can allow for the t_{2g} orbitals to localize and form low lying semi-core manifold well below the e_g orbitals [8]. We then can compare total energies for different scenarios corresponding to localizing a particular e_g orbital. By doing so one breaks the cubic symmetry but this is allowed if the resulting ground state is lower in energy. As a result of the orbital ordering the system will distort in order to reduce the electrostatic energy due to the interaction of the oxygen electronic clouds with the lobes of the e_g orbital, that is directed to them, on neighbouring Mn ions.

The basis states are written: $|x\rangle = x^2 - y^2$ and $|z\rangle = \frac{1}{\sqrt{3}}(2z^2 - x^2 - y^2)$. A composite state can be written as: $|\theta\rangle = \cos \frac{\theta}{2}|z\rangle + \sin \frac{\theta}{2}|x\rangle$ [9]. Then the orbital state $|z\rangle$ corresponds to $\theta = 0$ and the state $|x\rangle$ to $\theta = \pi$. The orbital ordering of LaMnO_3 consists of an antiferro ordering of two orbitals, namely $|\pm \theta\rangle = \cos \frac{\theta}{2}|z\rangle \pm \sin \frac{\theta}{2}|x\rangle$ in a plane while the same order is repeated along the third direction. Until recently it was assumed that $\theta = 2\pi/3$. But recent ESR [10] and neutron diffraction [11] measurements have estimated θ to be 92° and 106° respectively.

Phenomenological superexchange calculations for the ground-state ordering have also given $\theta_{opt} \sim 83^\circ$, ‘significantly different from $2\pi/3$ ’ [9]. Our current calculations are however limited to the cases of ferro order of $\theta = 0$ and π and antiferro order of $\pm\pi/3$ and $\pm 2\pi/3$.

2. Calculation details

The calculations are performed in the SIC-LSD approximation implemented within the LMTO-ASA method [12, 13]. The SIC corrects for the spurious interaction of an electron with itself inherent to the LDA approximation to the exchange correlation functional of the DFT. It is known however that this energy is important only when the electron is localized whereas it vanishes for delocalized electrons. This method is used to determine whether it is favourable for an electron to localize or to be itinerant. This is done by comparing the total energies of the system in the presence of the two scenarios. The lattice parameter used in the present calculation, $a_0 = 7.434$ au, is the one which gives the experimental volume of the real distorted LaMnO₃ system. We have used a minimal basis set consisting of $6s$, $5p$, $5d$ and $4f$ for La, $4s$, $4p$ and $3d$ for Mn and $2s$, $2p$, and $3d$ for O. Mn $4p$ and O $3d$ were downfolded. For the atomic sphere radii we used 4.01, 2.49 and 1.83 au for La, Mn and O respectively. In order to look at different orientations of the two orthogonal e_g orbitals we used rotations of the local axes on the Mn sites. We checked the accuracy of these rotations by comparing the total energies of three configurations: all $3z^2 - r^2$, all $3x^2 - r^2$ and all $3y^2 - r^2$ localized in both FM and G-AFM cases because these magnetic orderings preserve the cubic symmetry and hence the energy should not be dependent on which orbit is localized so long as it is the same one on all the Mn sites. The energy differences found in this way were always less than 1 mRyd per formula unit (fu).

The calculations were done for a four-formula unit cell. The notations of the orbital ordering scenarios are as follows: *lsd*: LDA calculation with no SIC; t_{2g} : SIC applied to the t_{2g} orbits only on all the Mn sites, and in all the other cases one e_g orbit is localized on top of the t_{2g} ones. The remaining scenarios correspond to localizing either the same or different orbits in the ab plane while preserving the same ordering on the second plane along c . Thus we have either ferro or C-type antiferro orbital ordering.

3. Results and discussion

From the total energies of table 1, we see that the ground state corresponds to an orbitally ordered solution forming a C-type antiferro-orbital arrangement of the $x^2 - z^2$ and $y^2 - z^2$ in the ab plane with the same ordering repeated along the c -axis. The corresponding magnetic ordering is of A-type AFM as found in the distorted system. This solution is however almost degenerate with the solution with an ordering of $3x^2 - r^2$ and $3y^2 - r^2$. The energy difference between the two solutions, 0.6 mRyd fu^{-1} , is within the accuracy of the calculation method (LMTO-ASA). It is then most likely that the true ground state of the cubic system is made up of a combination of both solutions. Interactions with the neighbouring oxygens are certainly different for the two orderings and relaxation of the oxygen positions in the real system may favour one of the solutions or a linear combination of them.

We have considered three types of spin order. Ferromagnetism and A type antiferromagnetism where the spins are parallel in the x - y -planes and the planes are stacked

Table 1. Total energies in mRyd per formula unit and magnetic moments in μ_B of cubic LaMnO₃ in the FM, A-AFM and G-AFM magnetic orderings with several orbital ordering scenarios. Where one orbit only is specified the orbital ordering is ferro and for two orbits e.g. $3x^2 - r^2/3y^2 - r^2$ the ordering is of a C-type with the ordering vector $\mathbf{q} = \frac{\pi}{a}(1, 1, 0)$. The energies are given as differences with respect to the energy of the solution corresponding to the experimentally assumed ordering of the distorted LaMnO₃.

Configuration		l_{sd}	t_{2g}	$3z^2 - r^2$	$x^2 - y^2$	$\frac{3x^2 - r^2}{3y^2 - r^2}$	$\frac{x^2 - z^2}{y^2 - z^2}$	$\frac{3x^2 - r^2}{3z^2 - r^2}$
Energy	FM	140.3	21.4	8.1	11.7	0.5	-0.5	6.3
	A-AFM	152.0	30.7	7.2	11.4	0.0	-0.6	4.9
	G-AFM	160.9	45.2	9.4	9.5	7.5	7.7	8.8
Mn mom.	FM	2.89	3.07	3.72	3.70	3.70	3.71	3.68
	A-AFM	2.81	3.14	3.62	3.69	3.70	3.67	3.67
	G-AFM	3.10	3.41	3.60	3.60	3.60	3.61	3.63

Table 2. Magnetic exchange constants in meV obtained from the total energies in table 1. J_1 and J_2 are Heisenberg in-plane and inter-plane exchange integrals respectively.

OO scenario	$3z^2 - r^2$	$x^2 - y^2$	$\frac{3x^2 - r^2}{3y^2 - r^2}$	$\frac{x^2 - z^2}{y^2 - z^2}$	$\frac{3x^2 - r^2}{3z^2 - r^2}$
$8J_1S$	14.96	-12.93	51.02	56.46	26.53
$4J_2S$	-6.12	-2.04	-3.40	-0.68	-9.52

antiparallel up the z -axis and G-type antiferromagnetism where each spin is antiparallel to all its neighbours. The difference in energy between the FM and A-AFM magnetic orderings in the two cases is also very small which is consistent with the fact that inter-plane AF exchange is much smaller than in-plane FM exchange in agreement with experiments. Experimental exchange integrals are obtained from fitting neutron scattering results (spin wave dispersion) to a simple Heisenberg Hamiltonian with two exchange integrals acting between nearest neighbours. We calculated the exchange constants using the convention of [14]: $E_F = (-4J_1 - 2J_2)S^2$, $E_{A-AF} = (-4J_1 + 2J_2)S^2$ and $E_{G-AF} = (4J_1 + 2J_2)S^2$ for the energies of the FM, A-AFM and G-AFM respectively. We assumed the value of $S = 2$ for the magnetic moment on Mn ions for all the orderings. The results are given, in table 2, for different orbital ordering (OO) scenarios of the e_g orbits as given in table 1. Experimentally the two exchange integrals are found to be $8J_1S = 13.36 \pm 0.18$ meV and $4J_2S = -4.84 \pm 0.22$ meV for the in-plane and inter-plane coupling respectively [14, 15]. We see then that our calculation overestimates the tendency to in-plane ferromagnetism whereas the interplane exchange is marginally underestimated. However it was found in LDA calculations [3] that the first neighbour exchange integrals depend dramatically on lattice distortions. This might explain why our exchange constants calculated for the cubic lattice are quantitatively different from the experimental ones which were determined for the distorted lattice. Our results are however in disagreement with recent model calculations of Sikora and Oleś [9] who found that for an ordering of ' $\theta = 2\pi/3$, often assumed for LaMnO₃,' the exchange constants 'are never close to the experiment'. Their calculated constants are both

ferromagnetic which contradicts the experimental fact that LaMnO_3 is an A-type antiferromagnet. Hence their argument that θ should in fact be different from the assumed $2\pi/3$.

The widely used Goodenough–Kanamori (G-K) rules [17, 18] give an indication of which exchange interactions should be positive (ferromagnetic) and which negative (antiferromagnetic) depending on the state of ionization of the two ions, the occupied orbitals and the angle subtended at the bridging ion. They are valid only for insulating states and were worked out using perturbation theory to give a general guide to the interactions although deviations are known to occur [19]. It is useful to compare our results for this specific material with the predictions of the G-K rules because the results may be used in future to assess the reliability of the rules. We note that in the case where we have ferromagnetism and ferromagnetically aligned orbits then our results predict a metallic ground-state and so in these cases the rules are not applicable.

In LaMnO_3 , the Mn ions are all in the same oxidation state and the Mn ions and the bridging oxygen lie along a straight line in the cubic unit cell; the bridging angle is π . Thus the only variable that is relevant to the G-K rules is the orbital order. The rules state that if nearest neighbour sites are occupied by the same orbit the interaction is negative, antiferromagnetic. The size of the effect depends directly on the overlap of the orbits e.g. if there are two orbits $3x^2 - r^2$ (which have large lobes in the x -direction) separated by a lattice vector directed along x it would be larger than if, for example, the two orbits were $3y^2 - r^2$ but still separated by a lattice vector directed along x . This would fit nicely with the result in table 2 that the value of J_2 (exchange up the z -direction) is negative for OO $3x^2 - r^2/3y^2 - r^2$ and also for OO $3x^2 - r^2/3z^2 - r^2$ but significantly larger in the latter case where there are $3z^2 - r^2$ orbits arranged in columns up the z -axis. The calculation of J_1 is more complicated [19] because the orbits are partially occupied but is ferromagnetic for OO $3x^2 - r^2/3y^2 - r^2$ [20]. The overlaps would be smaller in the x - y -plane for the case OO $3x^2 - r^2/3z^2 - r^2$ than for OO $3x^2 - r^2/3y^2 - r^2$ so application of the G-K rules would predict a larger value of J_1 (exchange in the x - y -plane) in the former case in agreement with first principles results. The signs of J_1 and J_2 in table 2 do agree with the G-K rules. There is one detail in which the first principles results do deviate from the G-K rules and that is in the case antiferromagnetic ordering with the ferromagnetic orbital order F $3z^2 - r^2$. In this case, since all the orbits are the same all the nearest neighbour interactions should be antiferromagnetic which would mean that the G-AF state should be more favourable than the A-AF state whereas the opposite order is seen in table 1 and the value for J_1 in table 2 should be negative for this orbit. The order is correct for the other ferromagnetic orbital order, F $x^2 - y^2$. Thus we see the predictions for the signs of J_1 and J_2 from the first principles calculation and the G-K rules agree in all cases (except the one mentioned for F $3z^2 - r^2$ orbit above) and the magnitudes agree. In one case, we see that there is a disagreement on the ordering of unfavourable states. Model perturbation calculations of the exchange constants also disagree with the G-K rules: as mentioned earlier, Sikora and Oleś [9] have found that for the case of $\theta = 2\pi/3$ the constants are small and both ferromagnetic, whereas G-K rules predict that J_1 is strongly ferromagnetic while J_2 is antiferromagnetic.

In the ferromagnetic case, the total moment is $4\mu_B$ which is the value one expects from having four d electrons. This is the case also because the FM solution is either half-metallic (OO scenarios $3z^2 - r^2$ and $x^2 - y^2$ of table 1) or insulating (OO scenarios $3x^2 - r^2/3y^2 - r^2$, $x^2 - z^2/y^2 - z^2$ and $3x^2 - r^2/3z^2 - r^2$ of table 1). The magnetic moment that is on the Mn ion can be less than this because of hybridization. The magnetic moment on the Mn ion when one e_g orbit is localized is about $3.70\mu_B$ in both FM and A-AFM solutions and of $3.60\mu_B$ in the G-AFM case. Because of hybridization with the oxygen part of the polarization is sitting on the oxygen ion.

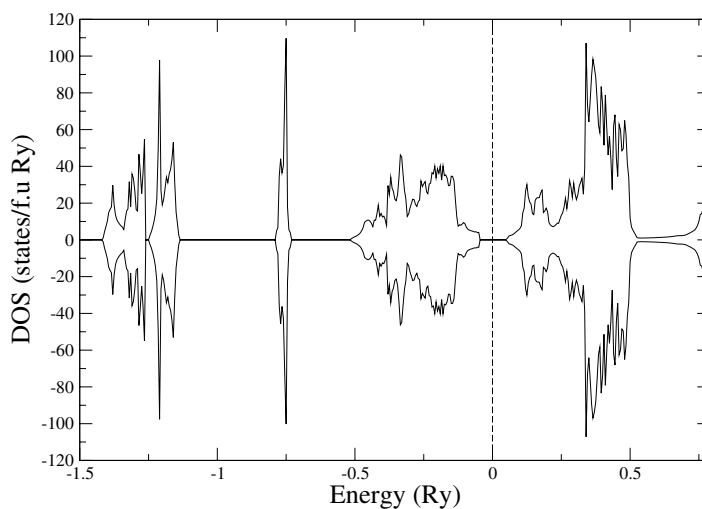


Figure 1. Total DOS in A-AFM magnetic and $x^2 - z^2/y^2 - z^2$ orbital orderings.

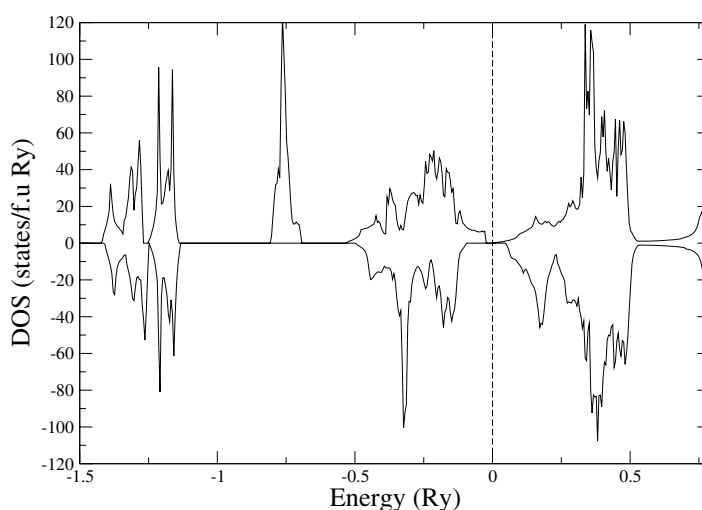


Figure 2. Total DOS in FM magnetic and $x^2 - z^2/y^2 - z^2$ orbital orderings.

The system is insulating in both orbital ordering scenarios independently of the magnetic ordering. Inspection of the total density of states (DOS) in the lowest energy $x^2 - z^2/y^2 - z^2$ ordering scenario presented in figures 1(A-AFM), 2(FM) and 3(G-AGM) reveals the presence of a gap which is larger as more nearest neighbour spins become antiferromagnetic (see also table 3). Its calculated value in the $3x^2 - r^2/3y^2 - r^2$ orbital and AFM magnetic orderings is in very good agreement with the experimental optical gap [21] as can be seen in table 3. The peak at about -0.75 Ryd in the total DOS corresponds to the localized $3t_{2g}$ and one e_g orbitals. The latter are shown in figure 4 where we can see the following features in the majority spin channel: the peak at -0.75 Ryd representing the localized $y^2 - z^2$ states and the $3x^2 - r^2$ states split into occupied states which hybridize strongly with the oxygen $2p$ states and unoccupied $3x^2 - r^2$ states. One can also notice by looking at the minority e_g states that both orbitals are degenerate because these are not corrected for by the SIC and hence are solutions of the LSD potential which

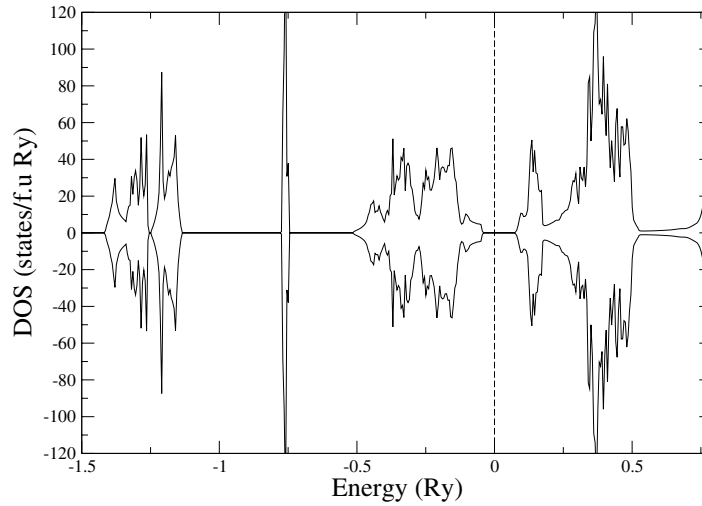


Figure 3. Total DOS in G-AFM and $x^2 - z^2/y^2 - z^2$ orbital orderings.

Table 3. Energy band gaps in eV.

Configuration	$3x^2 - r^2/3y^2 - r^2$	$x^2 - z^2/y^2 - z^2$	Exp
FM	0.54	0.27	
A-AFM	1.09	1.29	1.1 ^a
G-AFM	1.50	1.56	

^a [21].

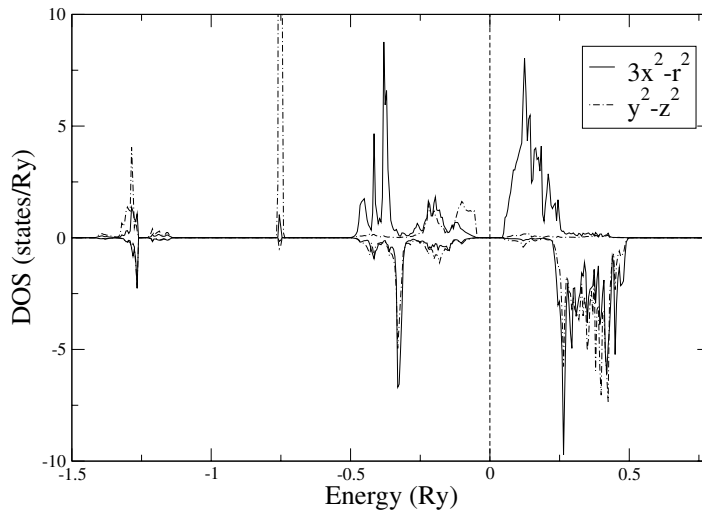


Figure 4. Mn e_g -projected DOS in the ground state A-AFM in the $x^2 - z^2/y^2 - z^2$ orbital ordering on the Mn site with the SIC applied to the $y^2 - z^2$ orbital.

are orthogonal to the SIC states. In the LSD calculation the t_{2g} and e_g states lie near the Fermi level with the t_{2g} states somewhat more localized than the e_g ones. However the LSD does not describe their localization accurately. In the SIC they are pushed well below the valence band,

composed mostly of oxygen $2p$ states. It is however known that the position of the SI-corrected levels does not correspond to what would be seen in experiment. Relaxation effects need to be considered if one wanted to get spectra from SIC single particle energies [16]. Centred around -1.25 Ryd are the oxygen $2s$ and La $5p$ semi-core levels.

The total energy of the solution where only t_{2g} orbitals are localized and the e_g electron is delocalized lies much higher than the most unfavourable orbital ordering solution which confirms that there is strong tendency to the localization of the e_g electron in LaMnO_3 even in the cubic phase. The energy scale of the localization/delocalization of the e_g electron is indeed at least twice as big as the energy corresponding to ordering the orbitals. This is qualitatively in agreement with the experimental observation that even above the critical temperature of the orbital ordering local distortions remain. Local distortions are an indication that there is localization. Once these e_g electrons are localized they induce local distortions through the interactions with the surrounding oxygens and these distortions order simultaneously with the orbitals when the temperature is lowered. Although we cannot with the current method simulate real paramagnetism as being a collection of disordered local moments without long range ordering we can speculate however, since the orbital ordering is so strong and independent of the spin ordering, that orbital ordering occurs in the paramagnetic state too. It is this orbital ordering which drives magnetic ordering and not the other way round. In a model calculation of paramagnetic LaMnO_3 and KCuF_3 based on an LDA + U electronic structure Medvedeva *et al* [22] concluded that distortions were not needed to stabilize the orbitally ordered phase in both compounds and that this ordering is of purely electronic origin. Their calculations for cubic LaMnO_3 have found that in the PM phase the orbitals order but they are not pure local $3z^2 - r^2$ and $x^2 - y^2$. They found that the local $3z^2 - r^2$ has an occupancy of 0.81 and the local $x^2 - y^2$ has an occupancy of 0.21. This is consistent with our present calculations in that the calculated ground state is nearly degenerate. In earlier LDA + U calculations [23] on KCuF_3 it was found that within LDA there was no instability of the system against distortion while in LDA + U the energy has a minimum for a finite distortion of the lattice. It was concluded then that electron-phonon and exchange only are not enough to drive the collective distortion. A similar view was supported also by model calculations [24, 25] where both electron-electron and electron-lattice interaction are taken into account. In our present calculation, the competition is rather in terms of localization/delocalization of the e_g orbitals by electronic interactions alone. And we found indeed that these are enough to first localize the orbitals (larger energy scale) and then to order them in an anti-ferromagnetic way (smaller energy scale). Based on these results and those mentioned earlier we speculate that the distortions are a consequence of the displacement of oxygen ions to accommodate the electrostatic interactions resulting from the orbital ordering but these are crucial in selecting the ground-state ordering out of the two nearly degenerate solutions we found for the cubic case.

Earlier SIC-LSD calculations by Tyer *et al* [26] have described correctly the physics of the distorted LaMnO_3 . Then Banach and Temmerman [27] studied the cubic phase but using a unit cell of two formula units only. This limited the study to the first two rows and first four columns of table 1. Hence they found that the lowest energy solution is the A-AFM with $3z^2 - r^2$ orbital ordering. Upon decreasing the lattice parameter they found a crossover to the FM with t_{2g} orbitals SI-corrected only which means suppression of orbital ordering. We reconsidered this case below with our present bigger cell.

Loa *et al* [28] studied structural and electronic properties of LaMnO_3 under pressure and found that the system is still insulating even at higher pressure than the critical one at which the structural transition takes place. There was no indication of the magnetic state of the system

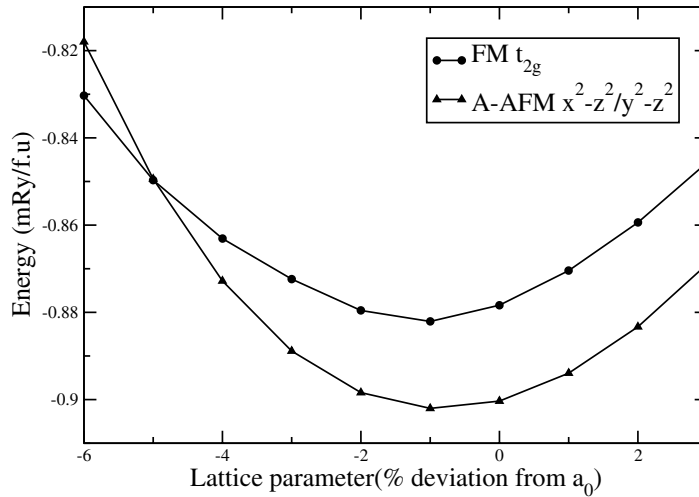


Figure 5. Total energies of the A-AFM $x^2 - z^2 / y^2 - z^2$ orbital ordering and FM t_{2g} solutions as functions of the deviation of the lattice parameter a from the experimental one $a_0 = 7.434$ au. We find a crossover from the e_g localized ordered state to the e_g delocalized as a is decreased.

but the experiments were carried out at room temperature which is well above the ordering temperature at least of the distorted LaMnO_3 . We found both FM and A-AFM solutions to be insulating in both $3x^2 - r^2 / 3y^2 - r^2$ and $x^2 - z^2 / y^2 - z^2$ orbital ordered states. Whereas the system is metallic when only the t_{2g} electrons are localized. The fact that the system was found to be insulating after suppression of the JT distortion is indicative of the presence of orbital ordering with or without spin ordering. Use of local probe such as EXAFS or pair distribution function (PDF) would be of great help though to settle the question of whether pressure really quenches distortions at the local level.

Another way of suppressing the distortions is by increasing temperature as done by Sánchez *et al* [29] who studied the structural changes of LaMnO_3 with temperature by using XANES and EXAFS measurements. Probing the local environment of the Mn ions they found no abrupt change in the signal upon crossing the structural transition temperature T_{JT} . They described the structural phase transition as ordering of the local distortions that are thermally disordered above T_{JT} resulting in a cubic lattice on average. This picture is quite different from the high pressure one although in both cases the distortions are apparently suppressed. In the high-temperature regime orbital ordering can still be present but the long range ordering is suppressed by thermal fluctuations. Consistent with our calculation that the localization/delocalization energy is of a larger scale than orbital ordering, i.e. the e_g electrons tend to localize strongly. As a consequence the lattice is distorted locally but since the energy scale of ordering the orbits/distortions is lower they are disordered by thermal fluctuations at high temperature.

We have also investigated the dependence of the orbital ordering on the volume of LaMnO_3 . To do so we compare total energies for different lattice parameters relative to the experimental one. The latter is determined by requiring that it gives the correct experimental volume of the distorted system. We compared the energies of two scenarios: the ground-state solution of the experimental volume ($x^2 - z^2 / y^2 - z^2$ orbital ordering and A-AFM spin ordering) and the FM solution with delocalized e_g orbits. The results are given in figure 5. One notices that the lattice

parameter corresponding to the minimum is the same in both solutions and that it is slightly smaller than the parameter obtained from the experimental volume of the distorted system. Upon decreasing the volume the two curves cross a at about -5% of the experimental lattice parameter. Below this value the e_g electron becomes delocalized and there is no longer orbital ordering. The system becomes metallic too as was signalled by the jump in the conductivity found by Loa *et al* [28].

4. Conclusions

We have investigated orbital ordering in cubic LaMnO_3 using the SIC-LSD method which allows to study the localization-delocalization competition of correlated electrons. Although orbital ordering in LaMnO_3 has been ascribed to Jahn–Teller distortions of the MnO_6 octahedra we found that this ordering can happen from purely electronic effects by spontaneous breaking of the cubic symmetry. Once the orbital ordering sets in the electrostatic interaction between the O ions and the electrons on the neighbouring Mn ions can be minimized by elongating the bonds along the lobes of the occupied e_g orbitals. It seems though that this coupling to the lattice is still needed to select the correct orbital ordering giving the observed distortions in the real LaMnO_3 system. There is therefore no need to assume an underlying A-AFM magnetic ordering to recover the orbital ordering. The latter is independent of the magnetic ordering and this is evidenced by the much higher ordering temperature of the orbits as compared to the spins. Although what we have found is that the lattice is important to determine the symmetry of the ground-state orbital ordering.

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