Das *et al.* Reply: The *first aspect* of the preceding Comment [1] by Markovich and Jung is the experimental controversy on the origin of the destabilization of charge ordering (CO) in $La_{0.5}Ca_{0.5}MnO_3$ nanoclusters. As rightly pointed out, the experimental synthesis of manganite clusters is faced with difficulties, e.g., impure phases, grain boundaries, and nonstoichiometry. Therefore, the experimental results reported by different research groups [2,3] may differ. Fortunately, our theoretical calculation [4] is devoid of these difficulties as we study a perfectly pure sample on the computer. Our calculation clearly shows that size reduction alone is able to destabilize CO in small $La_{0.5}Ca_{0.5}MnO_3$ nanoclusters. Nonstoichiometry, which is difficult to control experimentally might further add to this destabilization.

As a *second aspect*, the authors of Ref. [1] claim our calculations [4] to be "based on doubtful structural and magnetic results," i.e., the experimental results of Ref. [2]. The authors completely misconceive the nature of our calculation. There is no input from any experiment. Based only on the atomic species involved, we have done *ab initio*, parameter-free density functional theory calculations, performing a complete geometrical optimization of nanoclusters. The theoretical optimization, carried out at zero temperature, shows structural changes involving (i) a reduction in volume and (ii) a change in orthorhombic strain from an highly anisotropic one in bulk to a nearly isotropic one on the nanoscale. The change (ii) indeed *is in agreement* with the experimental findings in Ref. [3] quoted by the Comment's authors [1]: "Despite the similar room-temperature structure, the low temperature structural data of nanoparticles differ significantly from that of the bulk." This low temperature phase is relevant for the CO and is the one to be compared with our theoretical structural relaxation, instead of the room-temperature data as Comment [1] refers to.

Having clarified that the criticism of the Comment [1] concerning our *theoretical* calculation is unfounded, let us turn to the *experimental* controversy concerning the unit volume change. The results from different experimental groups [2,3] differ in this respect. While Ref. [3] reports a unit cell volume of 225 $Å^3$ compared to 224–225 $Å^3$ for the bulk, Ref. [2] reports a volume of 222 $Å^3$, i.e, a noteworthy reduction upon moving to the nanoscale. Our theoretical calculations predict a trend in volume change which is in agreement with that of Ref. [2] and with the general trend seen in theoretical and experimental studies in different systems [5] that the bonding becomes stronger as size decreases. Note that the amount of volume reduction strongly depends on the size of the nanoclusters. This is supported by our calculations: for 2 nm radius the volume reduction is 8% while for 3 nm radius it is 6%, in line with the experimental observation of 2% volume reduction for 15 nm clusters reported in Ref. [2]. For the larger 25 nm nanoclusters studied in Ref. [3], the general trend would suggest an even smaller volume reduction of 1% or less. This small change is challenging to resolve experimentally and prone to difficulties in the experimental synthesis procedure. On the other hand, the experimental study in Ref. [6], carried out on a related manganite, $Pr_{0.5}Ca_{0.5}MnO_3$, reports a systematic unit cell reduction when the particle size is reduced. A small to medium amount of volume reductions have been also reported in other works.[7]

We checked the validity of the predicted ferromagnetic phase to a larger cluster, by carrying out calculations with both the [2,4] 6% and 2% reduced volume, keeping the tilt and rotation of the MnO_6 octahedra the same as that found in our theoretical optimization [4]. The calculations showed the ferromagnetic state to be more stable compared to the antiferromagnetic CO state by 25 meV/f.u and 10 meV/f.u. for 6% and 2% volume reduction, respectively.

In conclusion, the only criticism of the Comment [1] concerning our *theoretical* calculations [4] is based on a misunderstanding as elaborated above. Our theoretical results [4] provide a clean stoichiometric system in isolation, devoid of the experimental problems. We unambiguously show that structural changes caused by the size reduction can lead to the destabilization of CO. The only point remaining is to clarify the contradicting *experimental* situation [2,3,6–8]. We are confident that future experiments, by studying smaller and systematically different sizes of nanoclusters grown under the same condition (as in Ref. [6]), will help to settle this issue.

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