Reply to "Comment on 'NMR in manganese perovskites: Detection of spatially varying electron states in domain walls' "

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In their Comment Savosta and Novák claim that the doubly peaked ^{55}Mn NMR line shape observed in $La_{0.67}Ca_{0.33}MnO_3$ for $T<70$ K [Phys. Rev. B **58**, 12 237 (1998)] is not real, but an artifact resulting from the experimental conditions. In support to their arguments, they present ^{55}Mn line-shape measurements at 77 K. In this reply we present ^{55}Mn NMR line-shape measurements under the experimental requirements suggested by Savosta and Novák, and show that at low temperatures the line shape still consists of two distinct peaks with different rf enhancement factors. This effect is explained in view of recent experimental results.

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In a recent paper[1] we have shown that at low temperatures the ^{55}Mn NMR spectra of $La_{0.67}Ca_{0.33}MnO_3$, obtained with a two-pulse spin-echo technique, are doubly peaked with peaks at frequencies 380 MHz and 395 MHz. This effect has been attributed to the lower charge carrier density in domain walls in comparison to the charge carrier density in ferromagnetic (FM) domains. For $T>70$ K, the double peak was no more observed, and spectra were found to consist of a broad single peaked Gaussian line shape.

In their Comment, Savosta and Novák claim that the double peak is due to the short spin-spin relaxation time T_2 value at the center of the line shape, which distorts the central part of the signal. They claim that by using very short interpulse time intervals τ, such that $\tau \ll T_2$, the double peak will disappear and the true single Gaussian line shape will be restored.

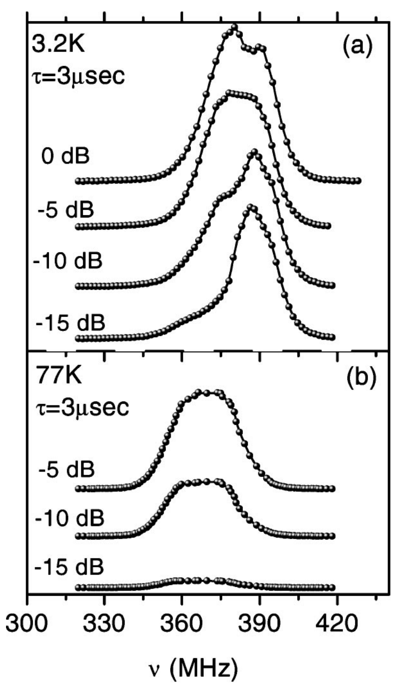

FIG. 1. (a) ^{55}Mn NMR line-shape measurements of $La_{0.67}Ca_{0.33}MnO_3$ at 3.2 K, and different rf power levels. (b) The same experiment at 77 K.

In order to clarify the question raised by them we performed low-temperature ^{55}Mn NMR line-shape measurements on $La_{0.67}Ca_{0.33}MnO_3$, after setting $\tau=3$ μsec, which in all measurements fulfilled the condition $\tau \ll T_2$. Figure 1(a) exhibits line-shape measurements at $T=3.2$ K, and at four different radio frequency (rf) power levels. It is clearly observed that by decreasing rf power the low-frequency part of the signal decreases rapidly, whereas the high-frequency part remains almost unchanged. This unambiguously shows that the signal consists of two components, which respond differently to the power level of the rf excitation pulse, i.e., they have different rf enhancement factor.[3] On the contrary, at 77 K [Fig. 1(b)] the line shape remains unchanged and only the signal intensity reduces drastically by decreasing rf power level, which implies a uniform rf enhancement across the whole spectum. The possibility that at $T=3.2$ K the doubly peaked line shape is produced by an extremely short T_2

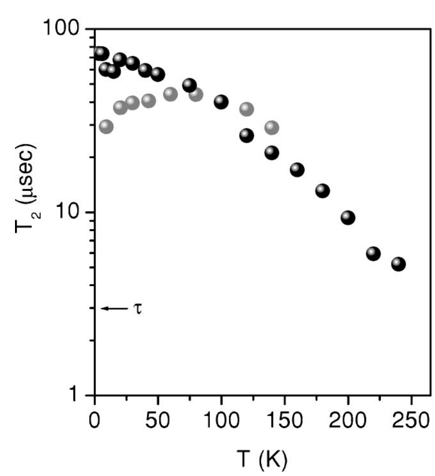

FIG. 2. ^{55}Mn T_2 as a function of temperature measured at rf power level -5 dB (filled circles). For reasons of comparison T_2 from Ref. 1 (gray circles) is also presented.

$\ll\tau$ at the center of the line shape, which distorts the line shape, is according to Fig. 2 excluded. In this figure we demonstrate T_2 vs T data for rf power -5 dB, obtained at the center of the line shape, where T_2 attains its minimum value. It is observed that by increasing temperature T_2 varies from 80 μsec$\gg\tau$ at 3.2 K to 5 μsec at $T=240$ K.[4] For reasons of comparison T_2 vs T data from Ref. 1 are also presented.

Conclusively, ^{55}Mn line-shape measurements in $La_{0.67}Ca_{0.33}MnO_3$ show clearly the formation of a bimodal distribution of frequencies with different rf enhancement at low temperatures. Since rf enhancement is produced by the coherent oscillation of electron spins in magnetic domains or domain walls,[2] it may be concluded that the two peaks are produced in spatially resolved FM regions with different electronic properties. The reason that in Ref. 1 this effect has been attributed to charge variation across the domain walls is (i) that different ^{55}Mn NMR frequencies reflect different Mn valence states, (ii) by applying an external magnetic field the high-frequency signal component reduces drastically, and (iii) T_2 varies continuously across the line shape. Clearly, a weak point of this model is that the saturation field in FM manganites is $H_{sat}<1$ T, as rightly indicated in the Comment, and signals from domain walls should not persist at higher fields. An alternative model might be the formation at low temperatures of a bimodal distribution of FM clusters with different charge carrier density. In such a case, the strong rf enhancement of the high-frequency peak would suggest the appearance of regions with reduced anisotropy, as the rf enhancement in FM domains is expressed by $n=H_{loc}/(H_{ext}+H_A)$, where H_{loc} is the local hyperfine field, and H_A the anisotropy field (in our case $H_{ext}=0$). Such a possibility sounds reasonable, especially after recent results,[5,6] which show the presence of orbitar ordering reorientation in the low-temperature FM phase of CMR manganites. It may also correlate with the rapid change from twofold symmetry to fourfold symmetry of the anisotropic magnetoresistance, observed to occur below 100 K in thin films of $La_{0.67}Ca_{0.33}MnO_3$.[7]

[1] G. Papavassiliou, M. Fardis, F. Milia, M. Pissas, G. Kallias, D. Niarchos, C. Dimitropoulos, and P. Scherrer, Phys. Rev. B **58**, 12 237 (1998).

[2] D. Weisman, L. J. Swartzendruber, and L. H. Bennett, in *Techniques of Metal Research*, edited by E. Passaglia (Wiley, New York, 1973), Vol. VI, and references therein.

[3] rf enhancement is the strong amplification of the applied radio frequency field H_1 at the nucleus, due to coupling with the electronic magnetization (Ref. 2).

[4] At higher power level, i.e., 0 dB as shown in Fig. 1, $T_2 > 80$ μsec.

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[7] J. O'Donnell, J. N. Eckstein, and M. S. Rzschowski, Appl. Phys. Lett. **76**, 218 (2000).