Response to ``Comment on `The controlled charge ordering and evidence of the metallic state in $Pr_{0.65}Ca_{0.35}MnO_3$ films' ''

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Response to "Comment on 'The controlled charge ordering and evidence of the metallic state in $Pr_{0.65}Ca_{0.35}MnO_3$ films' "

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Abstract

In a recent comment (Prellier W and Raveau B 2001 *J. Phys.: Condens. Matter* **13** 2749) on our published letter (Lee Y P *et al* 2000 *J. Phys.: Condens. Matter* **12** L133), have proposed that our $Pr_{0.65}Ca_{0.35}MnO_3$ films may have ($hk0$)-oriented epitaxy rather than the ($00l$)-oriented epitaxy. We point out herein that always, owing to the substrate–film lattice mismatch, lattice strains are accumulated during the heteroepitaxial growth of perovskite-like manganite films, which allows us to explain the physical results obtained.

In their Comment [1] on our Letter to the Editor [2], Prellier and Raveau have pointed out that the $Pr_{0.65}Ca_{0.35}MnO_3$ films prepared by us may have ($hk0$)-oriented epitaxy rather than the ($00l$)-oriented epitaxy. In their main argument they referred to electron diffraction data which were obtained for $Pr_{0.5}Ca_{0.5}MnO_3$ films [3]. Unfortunately, most publications give the opposite results for the manganite films grown on the $LaAlO_3$(001) substrate and show that the ($00l$) orientation of the films is realized (see, for example, references [4–10]). The low substrate temperature (≈600 °C) used in reference [3] might lead to such an uncommon result. The investigations carried out on the $Pr_{0.67}Sr_{0.33}MnO_3$ films show that a reduction in the substrate temperature to below 730 °C greatly degrades the microstructure and magnetic and transport characteristics of the film [4]. According to the Stranski–Krastanov mechanism for the nucleation and growth of the perovskite-like oxide films (initial layer growth followed by island growth), formation of the ($hk0$) texture is in principle unrealizable.

We agree with the comment that the three weak reflections ((103), (113) and (312)) are insufficient for a reliable calculation of a and b. However, the parameter c was obtained with great accuracy using the Nelson–Riley function for the treatment of x-ray diffraction data [2]. Because in our case $c = 0.774$ nm and 0.7719 nm for the as-deposited and annealed films,

respectively, and $c = 0.7671$ nm for bulk $Pr_{0.65}Ca_{0.35}MnO_3$ [11], we came to the conclusion that the crystal lattice of the film is different from that of the bulk material.

Accumulation of lattice strains during the heteroepitaxial growth of manganite films is a well-known and well-studied phenomenon [4, 12, 13]. The change of Mn–Mn interatomic distances, and consequently the Mn–O–Mn bond angles, induced by the misfit strain can lead to significant changes in the magnetic and electronic properties of the perovskite manganites [12–14]. Therefore, the observation of charge ordering in the epitaxial $Pr_{0.65}Ca_{0.35}MnO_3$ film controlled by the lattice strains is not unexpected.

In addition we present the magnetization curves $M(T)$ obtained for the as-deposited and annealed $Pr_{0.65}Ca_{0.35}MnO_3$ films investigated in our Letter to the Editor [2]. Figure 1 shows that the magnetization value of the as-deposited film is 1.5 times that of the annealed one. Consequently, the as-deposited film contains more ferromagnetic (and metal-like) phase than the annealed one. Moreover, an increase in the annealing time up to 10 h does not change the $M(T)$ behaviour, which is directly correlated with the behaviour of the out-of-plane crystal lattice parameter [2]. For comparison, we include the $M(T)$-versus-T curve for $Pr_{0.65}Ca_{0.35}MnO_3$ film deposited onto a YSZ(001) substrate under similar conditions. Because the lattice parameter of YSZ is $a = 0.5142$ nm, the lattice mismatch between the substrate and film is about 5.6%, which is four times that for the $LaAlO_3$ substrate. It is seen that the increase of the lattice strains results not only in the growth of the magnetization value, but also in an increase of the Curie value, which is typical for manganite films under the action of compressive pressure [15].

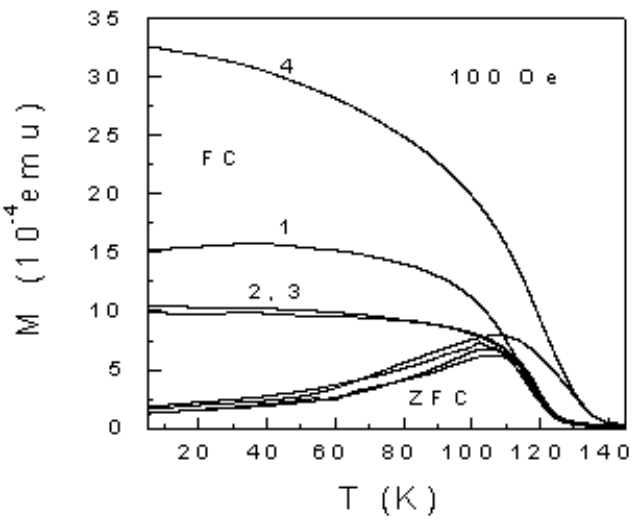

Figure 1. Temperature dependences of field-cooled (FC) and zero-field-cooled (ZFC) in-plane magnetizations under 100 Oe for $Pr_{0.65}Ca_{0.35}MnO_3$ films: (1) as-deposited on LAO, (2) after annealing for 1 h at 900 °C, (3) after annealing for 10 h at 900 °C, and (4) as-deposited on YSZ.

In summary, we have pointed out that *always*, owing to the substrate–film lattice mismatch, lattice strains are accumulated during the *heteroepitaxial* growth of perovskite-like manganite films. Their presence in epitaxial $Pr_{0.65}Ca_{0.35}MnO_3$ films allows us to explain the physical results obtained, including the controlled charge-ordering phenomenon and the appearance of the metal-like state. In addition, all of these peculiarities in the physical properties can be observed only in well-prepared *epitaxial* films with a correct orientation of the film texture.

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