Comment on "Reflection High-Energy Diffraction Oscillations during Epitaxial Growth of High-Temperature Superconducting Oxides"

Using x-ray diffraction (XRD) and reflection high energy electron diffraction (RHEED), Terashima *et al.* [1] attempted to demonstrate that RHEED oscillations correspond to layer-by-layer growth of a $YBa_2Cu_3O_7$ (YBCO) thin film. We have reanalyzed their experimental data and find serious objections to their analysis. Therefore, the growth of a single, segregated unit cell YBCO film has not been proven and hence the issue of the superconductivity of a single unit cell remains an open question.

Elementary diffraction theory implies that a film of finite thickness exhibits broadened Bragg peaks and finite size diffraction peaks. The film thickness L can be estimated from the Scherrer formula [2]

$$L=0.9\lambda/[\Delta(2\theta)\cos\theta_0], \tag{1}$$

where λ is the x-ray wavelength and $\Delta(2\theta)$ is the full width half maximum of the Bragg peak centered at θ_0 [3]. Alternatively, L may be determined from the finite size peak-to-peak or trough-to-trough distances using

$$L=1/2\lambda/(\sin\theta_n-\sin\theta_{n-1}), \tag{2}$$

where θ_n, θ_{n-1} are the positions of two adjacent finite size peaks or troughs. Figure 1 displays the results for both methods applied to the XRD spectrum of the YBCO film from Ref. [1]. The thickness measurements range from 39 to 65 Å with a mean of 51 Å (4.4 unit cells) and a standard deviation of 9 Å. Terashima *et al.* claim the thickness to be "in good agreement with 5 times the lattice spacing" (58.5 Å). Clearly in the work of Terashima *et al.* there is a large discrepancy in the mean, in addition to an 18% standard deviation. Moreover, their (005) peak width and thickness obtained presumably through Eq. (1) are inconsistent. As a result, the XRD data disagree with the claim of exactly 5 unit cells thickness and thereby does not imply layer-by-layer growth.

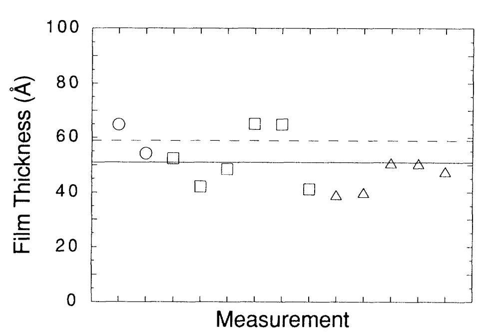

FIG. 1. Film thickness derived from Fig. 2(c) of Ref. [1], using Scherrer formula (○), trough-to-trough (□), and peak-to-peak (△). The solid and dashed lines indicate the mean and the value claimed by Terashima *et al.*, respectively.

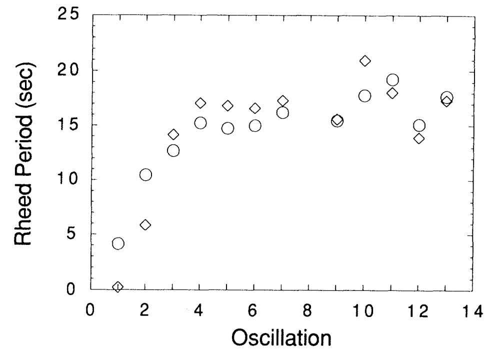

FIG. 2. RHEED period vs oscillation derived from Fig. 2(c) of Ref. [1], using peak-to-peak (○) and twice peak-to-trough (◇) measurements.

Recently, layer-by-layer growth as characterized by RHEED oscillations has been questioned [4], theoretically using Monte Carlo simulations [5], and experimentally by comparing to scanning tunneling microscopy (STM) [6]. In any case, assuming that RHEED oscillations indicate layer-by-layer growth, the period exhibits large variations as shown in Fig. 2. In order to compare to the XRD data in the "numbering of the RHEED oscillations peaks," Terashima *et al.* "neglected the initial transient peak." Of course, this very first peak is the most important one when the physics of a single layer is studied. Clearly, large systematic and random errors exist in the period determination using these RHEED oscillations. Finally, it should be stressed that RHEED or qualitative analysis of XRD cannot address the quantitative issue of interdiffusion.

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