Reflection high-energy electron-diffraction studies of the growth of $YBa_2Cu_3O_{7-x}$ and $DyBa_2Cu_3O_{7-x}$ superconducting thin films

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Studies of intensity oscillations of the specularly reflected reflection high-energy electron-diffraction (RHEED) beam during the growth of $YBa_2Cu_3O_{7-x}$ and $DyBa_2Cu_3O_{7-x}$ thin films on (100)$SrTiO_3$ substrates are presented. These studies have been conducted under controlled conditions of atomic fluxes, substrate temperatures, and oxygen partial pressures. The periods of the oscillations have been observed to increase when the substrate vicinal angle is increased, at fixed flux. Oscillation amplitudes have been found to depend on the substrate temperature. The oscillation time periods as well as amplitudes for the Dy and Y compounds have been found to be dramatically different. These observations have been characterized within the framework of a model of RHEED oscillations, which attributes them to changing surface step densities and distributions. After appropriate normalization, the time dependences of the RHEED oscillations obtained at different temperatures coincide for each compound. This indicates that these oscillations are a consequence of surface steps. Such an interpretation yields qualitatively accurate predictions of observed intensity oscillation profiles and is also consistent with transmission-electron-microscope (TEM) studies of nucleation and growth. Bare substrate areas have been found in TEM studies in films which, during growth, exhibited 12 periods of RHEED intensity oscillations. Discrepancies have been found in the thicknesses of films as measured by RHEED oscillations and Rutherford backscattering. These results indicate that assertions of superconductivity in single unit-cell thick layers based on the observation of RHEED oscillations are open to question.

I. INTRODUCTION

Reflection high-energy electron diffraction (RHEED) is well suited to the *in situ* study of epitaxial growth and of the evolution of surface morphology. At the low grazing angles commonly used, RHEED is exceedingly surface sensitive, capable of resolving order over long distances. The low incidence angle also reduces the Debye-Waller factor and makes RHEED compatible with moderately high growth temperatures. Intensity oscillations of the specularly reflected RHEED beam during molecular beam epitaxy (MBE) have been used to calibrate fluxes and measure growth rates.[1-4] There are two approaches to the origin of these intensity oscillations. Neave *et al.*[1] have suggested that they are a consequence of diffuse scattering from step edges. Lent and Cohen[5] have developed a kinematic approach, where the intensity oscillations are derived from Bragg-type interference of beams scattered by terraces which are at two levels, a monolayer apart. In both models, a description of the observed oscillations requires expressions relating the time evolution of the step density and/or terrace areas with experimental parameters and material constants of the crystallizing substance.[6]

Terashima *et al.*[4] have reported studies of oscillations in the intensity of the specular RHEED beam during the growth of $YBa_2Cu_3O_{7-x}$ films on (100)$SrTiO_3$ substrates. These oscillations have been interpreted to indicate two-dimensional layer-by-layer growth, with each layer of the film being one unit cell high, thereby leading to the possibility of fabricating multilayers and artificial structures to study superconducting behavior in reduced dimensions. The nucleation of the film has been considered to occur on the terraces, which separate the steps. Based on this work, Terashima *et al.*[4] have reported superconductivity in single unit-cell layers of $YBa_2Cu_3O_{7-x}$. This report of layer-by-layer growth is in contradiction with studies of nucleation and growth of $YBa_2Cu_3O_{7-x}$ reported in the literature,[7-9] where imaging techniques indicate nucleation at step edges. It should be mentioned here that there exist certain differences in the film and substrate materials and deposition processes in each of these studies. For example, while Terashima *et al.*[4] have studied $YBa_2Cu_3O_{7-x}$ films on (100)$SrTiO_3$ grown by reactive coevaporation, Streiffer *et al.*[7] have studied $YBa_2Cu_3O_{7-x}$ films on (100)MgO grown by sputtering, while Agrawal *et al.*[9] have studied $DyBa_2Cu_3O_{7-x}$ films on (100)$SrTiO_3$.

It is the purpose of this work to address these discrepancies and study the growth of $YBa_2Cu_3O_{7-x}$ and $DyBa_2Cu_3O_{7-x}$ films on (100)$SrTiO_3$ under controlled experimental conditions of atom fluxes, substrate temperatures, and oxygen partial pressures, using reactive coevaporation as the growth process. It has also been our intention to minimize effects due to substrate surface structure. Precautions such as the use of substrates from the same lot, from the same wafer, and from one vendor, have been taken.

II. EXPERIMENTAL RESULTS

All films were grown on (100)$SrTiO_3$ using an ozone-assisted, reactive coevaporation process, identical to that of Terashima *et al.*[4] The cation fluxes were carefully monitored and kept identical (to within the experimental resolution of the quartz crystal monitors used to measure the fluxes) during all experiments. Furthermore, the substrate temperature and the ozone inlet pressure (which was monitored using a capacitance manometer) were both maintained constant. This ensured a controlled environment at the substrate surface so that any observations of RHEED intensity oscillations could be attributed to the response of the surface to nucleation and growth of the film. Two sets of vicinal substrates of $SrTiO_3$ were also used. These were 2.2° and 4.4° off (100) towards [110]. These substrates were annealed at 1250 °C in air for a period of 1 h in order to induce surface reconstruction.[9] The deposition parameters were maintained the same for these vicinal substrates.

The RHEED studies were carried out during growth at an accelerating voltage of 10 kV and an incident angle of 28 mrad (1.6°) as determined from the system and deposition geometry. The electron beam was incident along the [100] crystallographic direction, and the intensity of the specularly reflected spot was monitored using a commercially available photomultiplier and current amplifier circuit. Further, the amplifier gain was kept constant during all studies in order to ensure that the absolute values of measured intensities have the same basis for meaningful comparison.

Figure 1(a) shows the intensity oscillations of the specularly reflected RHEED beam, at a Bragg diffraction condition, observed during the growth of a $DyBa_2Cu_3O_{7-x}$ film on a normal (100)$SrTiO_3$ substrate. At the Bragg condition, the diffraction is least sensitive to the presence of surface steps.[10] This film was grown under deposition conditions favoring *c* normal growth (the *c* axis of the film being normal to the substrate surface). The time period of the oscillations was about 8.4 s. The cation deposition rates were maintained so as to yield a film growth rate of 1.07 Å/s. Ten oscillations are clearly visible. It is of interest to note the damping of the oscillations. This has important consequences, which will be discussed later. Figure 1(b) shows the effect of annealing the film at the deposition temperature, with the ozone flow held constant. During annealing, the RHEED specular intensity is seen to build up, however, it never reaches its original value. The thickness of the film as calculated from the growth rate is 1920 Å. Rutherford backscattering yielded 2000 Å. An evaluation of the thickness, considering each RHEED oscillation as a signature of the completion of one unit cell, yielded 2500 Å, a 25% discrepancy. Clearly there is no relation between the number of RHEED intensity oscillations and film thickness.

In Figs. 2(a) and 2(b), the effect of a slight change in substrate orientation on the RHEED intensity oscillations is presented. Figure 2(a) is a plot of oscillations during the growth of a $DyBa_2Cu_3O_{7-x}$ film on a reconstructed 2.2° off (100)$SrTiO_3$ substrate, and Fig. 2(b) is for growth on a 4.4° off (100)$SrTiO_3$ substrate. It should be noted here that the deposition conditions have been kept identical to the previous case, i.e., the cation fluxes (atoms/cm^2/s) are the same, the substrate temperature is kept constant, as is the ozone flow. These oscillations were also obtained at a Bragg diffraction condition, along a [100] azimuth. As the vicinal angle is increased, the distinctly different response of the surface to the nucleation and growth of the films is readily apparent, in terms of enhanced damping, as well as an increase in the time period of the intensity oscillation itself. These results demonstrate differences in the growth morphology on vicinal surfaces of different angles. The streaks observed during the growth on vicinal surfaces appeared finer and longer than during the growth on normal substrates, as shown in Fig. 3, presumably indicating better

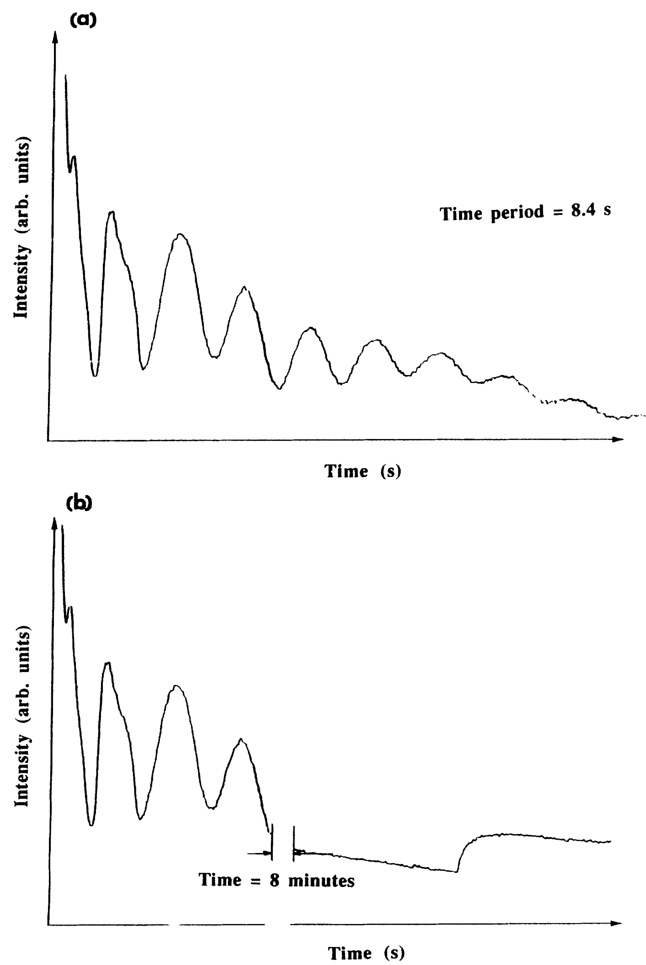

FIG. 1. (a) Oscillations in the intensity of the specular RHEED spot during the growth of a $DyBa_2Cu_3O_{7-x}$ film on a (100)$SrTiO_3$ substrate. The beam was incident along [100] at a Bragg condition and the substrate temperature was maintained at 760 °C. The oscillation time period is 8.4 s. (b) Effect of annealing the substrate at the deposition temperature in flowing ozone. The RHEED intensity builds up gradually, suggesting diffusional smoothening of the surface. However, the intensity never reaches its original value.

film morphology, consistent with published results.[11]

The oscillations of the specular beam intensity, observed during the growth of a $YBa_2Cu_3O_{7-x}$ film on a $(100)SrTiO_3$ substrate, maintaining the same deposition conditions except for the change of rare earth, are shown in Fig. 4. The rare-earth flux has been kept the same as that in Fig. 3 where Dy was used. The increase in the time period and enhanced damping (in comparison to $DyBa_2Cu_3O_{7-x}$) are readily apparent. Variation of substrate temperature appears to have an effect qualitatively similar to that observed in GaAs MBE,[12,13] i.e., an increase in temperature appears to reduce the RHEED oscillation amplitude and increase the damping. This has been observed for both $YBa_2Cu_3O_{7-x}$ as well as $DyBa_2Cu_{7-x}$ films. This effect, however, is not quite a monotonic function of temperature in this system, in contrast to GaAs MBE.

At the off-Bragg condition (where the diffraction is most sensitive to the presence of steps), we generally observe an initial increase in the intensity. This is illustrated in Fig. 5, which shows RHEED oscillations observed during the growth of $YBa_2Cu_3O_{7-x}$ film, where the diffraction condition is off Bragg. This increase, however, is soon damped out. No change in the oscillation time period has been observed relative to the Bragg condition. In general, a larger number of oscillations are seen at the off-Bragg condition. In contrast to the observations of Terashima *et al.*,[4] we do not observe any transient with a

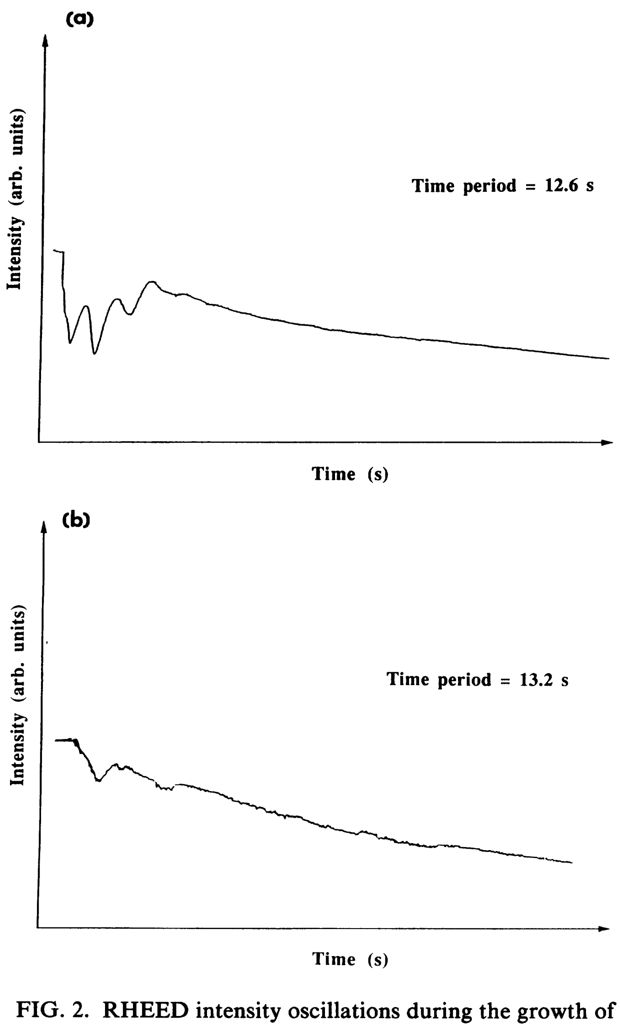

FIG. 2. RHEED intensity oscillations during the growth of a $DyBa_2Cu_3O_{7-x}$ film on vicinal $(100)SrTiO_3$ substrates. (a) 2.2° off (100) towards [110] and (b) 4.4° off (100) towards [110]. The surface steps are along [100]. Here, the incident beam is both parallel and perpendicular to the steps. The increased time period (relative to Fig. 1) and enhanced damping are readily apparent.

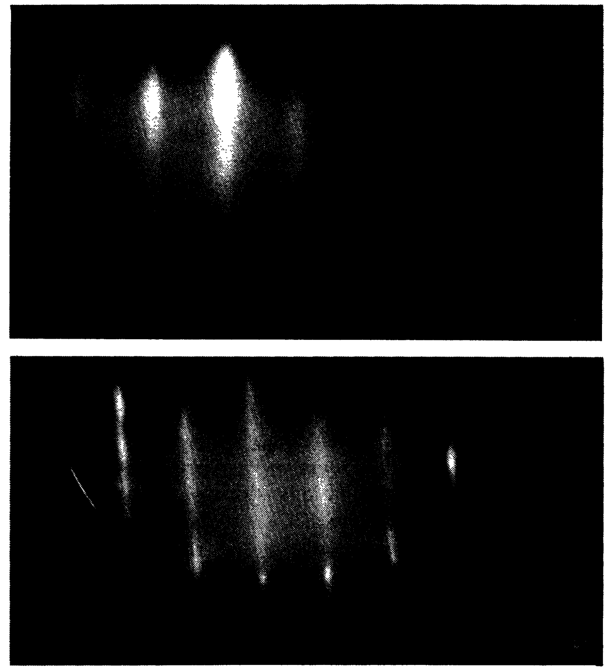

FIG. 3. RHEED patterns during the growth of a $DyBa_2Cu_3O_{7-x}$ film on (a) normal and (b) vicinal 2.2° off $(100)SrTiO_3$ substrates. The finer streaks in (b) are indicative of better film morphology.

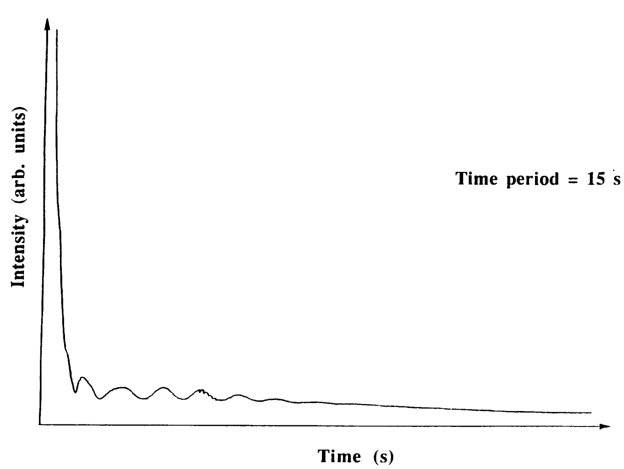

FIG. 4. Intensity oscillations during the growth of a $YBa_2Cu_3O_{7-x}$ film on a $(100)SrTiO_3$ substrate, the experimental parameters being the same as in Fig. 1(a). The enhanced damping and increased time period (15 s) are noteworthy.

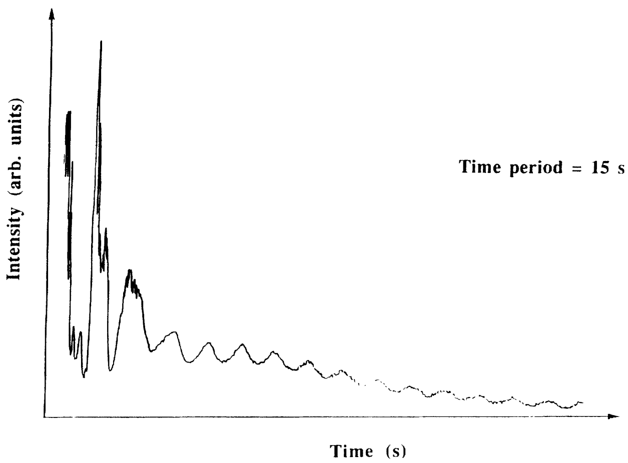

FIG. 5. Intensity oscillations during the growth of a $YBa_2Cu_3O_{7-x}$ film on a (100)$SrTiO_3$ substrate, the experimental parameters being the same as in Fig. 1(a), the diffraction condition is off Bragg.

time period different from the subsequent oscillations. The observations described here are relevant to $YBa_2Cu_3O_{7-x}$ as well as to $DyBa_2Cu_3O_{7-x}$ films.

III. POSSIBLE ORIGINS OF RHEED INTENSITY OSCILLATIONS

At the outset, we wish to point out an important difference between MBE growth in the film-substrate system studied here and that involving both Si and GaAs. The system we study here is predominantly ionic, with some degree of covalency, in contrast to Si and GaAs which are entirely covalent. This has certain fundamental consequences for the nature of epitaxial growth in these systems. An important consequence of the ionicity is the requirement of charge neutrality, i.e., as the film grows, it is required to be electrically neutral. This requirement leads to unit-cell by unit-cell growth, since one unit cell would satisfy the charge neutrality requirement. Streiffer *et al.*[7] have suggested one unit cell to be the growth unit, the growth taking place by simultaneous attachment of all species on the *a*-*c* or the *b*-*c* planes, and resulting in the propagation of a one-unit-cell-high ledge of $YBa_2Cu_3O_{7-x}$.

It is important to initially consider the substrate surface structure itself since it is generally accepted that this is one of the important factors influencing growth. The presence of steps on surfaces influencing nucleation is well documented.[7–9] $SrTiO_3$ surfaces can have two possible terminations,[14] i.e., Sr or Ti. Naively, this would lead one to expect steps on $SrTiO_3$ surfaces to be a half unit cell high. This is to be contrasted with the growth unit of $YBa_2Cu_3O_{7-x}$, which has been mentioned to be one unit-cell high. Most treatments of the origin of RHEED intensity oscillations[1–3] consider systems in which the steps on both the film and substrate are one or two monolayers apart. Important differences in growth mechanisms are to be expected for these two cases. Further, the dissimilarity in the crystal structures of the film and substrate materials leads to two epitaxial relationships, which have been found to be determined during the initial stages of epitaxial growth itself.[9]

Large differences in the step heights of the film and substrate would naturally mask the effect of the presence of substrate surface steps, leading to the postulation of two-dimensional island nucleation and growth on terraces.[4] The electron beam, with a de Broglie wavelength of 0.1 Å, will be scattered quite strongly by steps that are an order of magnitude longer (substrate steps) and even more strongly by film steps. Two crystallographic variants of growth have been observed during the growth of $YBa_2Cu_3O_{7-x}$ and $DyBa_2Cu_3O_{7-x}$ films, namely, *c* normal and *a* normal. These have been observed to occur in different temperature regimes, thereby establishing the influence of species supersaturation at the substrate surface on the epitaxial relationship.[9]

The growth mechanism results in steps of different heights for *c* normal and *a* normal regions of the film. In contrast to *c* normal growth *a* normal growth results in steps that are 3.9 Å high, since this is the relevant dimension of the unit cell. These steps are merely a factor of 2 higher than the substrate steps in contrast to steps for *c* normal growth, which are a factor of 6 higher. Differing rates of growth along either the *a* or *c* crystallographic directions of the film, coupled with the differences in step heights, lead to dramatically different RHEED intensity oscillations for these two cases. Further, the time period of the oscillations for *a* normal growth is not a third of that for *c* normal growth, which is what would be expected based on the unit-cell dimensions, at constant cation fluxes, assuming a growth rate independent of crystallographic orientation. This observation is clearly in contradiction with the experimental data, which shows that growth along the *a* or *b* crystallographic directions is faster than growth along the *c* direction. The larger time period oscillations are damped out faster.

Stoyanov and Michailov[6] have found that the time evolution of the terrace areas (in the case of complete condensation, such as MBE) is temperature independent, which leads to a temperature-independent amplitude of the RHEED intensity oscillations, when they originate from the interference of beams scattered by different terraces.[5] When the RHEED intensity oscillations originate from diffuse scattering from the step edges, the oscillation amplitudes observed at different temperatures are expected to be essentially different, but the damped oscillations should coincide after appropriate normalization.[6] In the next section we discuss evolution of the surface step densities, accounting for the step height differences (by using the step height itself as weighting factor) following Stoyanov and Michailov.[6]

IV. EVOLUTION OF SURFACE STEP DENSITIES

A certain reference step height is required for the evaluation of the surface step density. In the case of Si and GaAs, this is quite naturally the height of the growth unit, a monolayer. In the case of epitaxial growth of dissimilar materials, it appears natural to use the smallest step height as the reference. The total surface step density may be evaluated by summing the contributions of the substrate and the film, after weighting each appropriately.

In the case of Si MBE growth, the amplitude of the oscillations changes negligibly in a time interval containing many oscillation periods,[3] in contrast to $YBa_2Cu_3O_{7-x}$ and $DyBa_2Cu_3O_{7-x}$. The assumption of conservation of the growth front thickness[6] may thus be applicable to the growth of Si or GaAs, but not to $YbA_2Cu_3O_{7-x}$ and $DyBa_2Cu_3O_{7-x}$. Conservation of growth front thickness leads to absence of damping in the RHEED oscillations, while the presence of damping in the latter is suggestive of thickening of the growth front. Thus, observations of strong damping in the RHEED oscillations in the growth of $YBa_2Cu_3O_{7-x}$ and $DyBa_2Cu_3O_{7-x}$ films suggests the presence of multiple growth levels on the surface.

Stoyanov and Michailov[6] have investigated the step density oscillations on a two-level and three-level growth front, using the Kolmogorov expression for the time evolution of the surface coverage,

$$\theta(t)=1-\exp\left[-\pi N_n\left[\int_0^t c(\tau)d\tau\right]^2\right] , \quad (1)$$

where θ is the crystallized fraction whose growth starts at $t=0$ with simultaneous appearance of N_n nuclei and the step velocity $c(\tau)$ is time dependent. Instead of monolayers, θ here pertains to the relevant growth front, which is a unit cell high. The step density at the mth level, $L_m(t)$, may be expressed as

$$L_m(t)=\frac{1}{c_m(t)}\frac{d\theta_m}{dt}=2\sqrt{\pi N_n}(1-\theta_m)\sqrt{-\ln(1-\theta_m)} \quad (2)$$

with the total step density being

$$L(t)=2\sqrt{\pi N_n}\sum_{m=1}(1-\theta_m)\sqrt{-\ln(1-\theta_m)} . \quad (3)$$

The time-dependent step velocities on consecutive levels $m-1$ and m are coupled by the diffusion field on the surface of the $(m-1)$th growth level. An essential feature of the above equations is the absence of any parameter containing material constants. This means that the coverage, and therefore the sum in Eq. (3), depends on neither experimental conditions nor material constants.[6] The evolving step density on the crystal surface depends on experimental parameters and material constants through the quantity N_n, the number of nuclei. This universality of the coverage is even more important when the RHEED intensity oscillations are due to the interference of beams scattered by terraces separated by elementary steps.[6]

Considering the general case of more than one growth level (in terms of unit cells), we note that, with the weighting factor of the step heights, the step density may be expressed as

$$L(t)=L(0)+2k\sqrt{\pi N_n}\sum_{m=1}(1-\theta_m)\sqrt{-\ln(1-\theta_m)} , \quad (4)$$

where $L(0)$ is the step density on the bare substrate. For the c normal growth case, the factor k is 6, and for the case of a normal growth k equals 2. These values arise from the difference in step heights of film and substrate. The amplitude of the step density oscillations is expected to increase rapidly to attain the value corresponding to the new material constants.

V. DISCUSSION

The strong damping of the intensity oscillations during the growth of the $YBa_2Cu_3O_{7-x}$ and $DyBa_2Cu_3O_{7-x}$ films is natural within the framework of the model discussed above. The damping appears stronger in the case of the former. This difference could arise due to the quantity N_n, could be larger for $YBa_2Cu_3O_{7-x}$ than for $DyBA_2Cu_3O_{7-x}$. The time period of the oscillations is also larger for $YBa_2Cu_3O_{7-x}$ than for $DyBa_2Cu_3O_{7-x}$. An estimation of the step velocity for the growth of $DyBa_2Cu_3O_{7-x}$ (Ref. 15) indicates that the step velocity increases with increasing vapor pressure. This appears reasonable since the vapor pressure of Dy is higher than that of Y. A higher step velocity also implies a faster oscillation in the surface step density, explaining the differening time periods.

For the case of a normal and c normal growth, the factor k is expected to play a role in influencing the number of oscillations that are seen. It follows from the model discussed above that more oscillations would be seen in the former case than in the latter. This is again found to correlate well with experimental observations, where we have seen a larger number of oscillations for the a normal growth, as shown in Fig. 6. Also, the damping of the first oscillation appears larger in the case of c normal growth, relative to a normal growth, consistent with the above interpretation.

For the case of vicinal surfaces, we expect that the specular spot intensity from the bare substrate would be significantly lower, in comparison to normal substrates. This fact is confirmed by Figs. 1 and 2, where, despite maintaining a constant gain on the photomultiplier amplifier, the starting intensity is seen to be lower for the

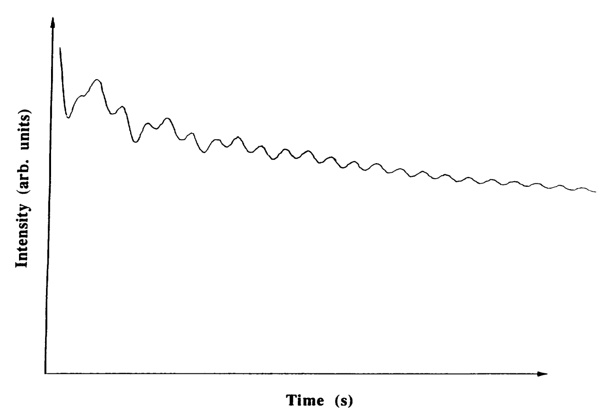

FIG. 6. RHEED intensity oscillations during the growth of an a-axis-oriented $DyBa_2Cu_3O_{7-x}$ film on (100)$SrTiO_3$. In comparison to Fig. 1, these are less damped, enabling the observation of a larger number of oscillations.

vicinal substrates by as much as 50%. Further, we expect fewer oscillations and increased time periods. The time period is expected to increase because we are distributing a fixed amount of material amongst an increased number of steps, thereby reducing the step velocity. The surface requires a larger time interval to reach its original step density. This is consistent with our experimental results. The time period of the oscillations is found to increase with reducing substrate temperature, for the same atom flux and major fraction of crystallographic orientation of the film being the same. This is indicative of a diffusion-controlled evolution of surface morphology, consistent with the interpretation of RHEED oscillations arising from diffuse scattering by steps.

We find that the time dependences of the RHEED intensity, obtained at different temperatures, coincide reasonably well after appropriate normalization. This is evident in Fig. 7, where, following Sakamoto *et al.*,[3] we plot the normalized intensity for $DyBa_2Cu_3O_{7-x}$ against the number of oscillations. The behavior of $YBa_2Cu_3O_{7-x}$ is quite different compared to $DyBa_2Cu_3O_{7-x}$, as illustrated by Fig. 8. The coincidence of the plots at different substrate temperatures indicates that the RHEED intensity oscillations are due to diffuse scattering from step edges and not due to layer-by-layer growth.

The fabrication of multilayered structures for the study of superconductivity requires a suitable insulating oxide, which not only has the right structure for the attainment of good epitaxial relations, but which is also compatible in terms of growth mechanisms. In Fig. 9, we compare the normalized RHEED oscillation intensities for rare-earth oxides which were grown on $(100)SrTiO_3$ substrates at identical conditions except for reduced atom fluxes, and the superconductors. The incompatibility of these oxides and superconductors for the growth of multilayered structures is readily apparent, since the slopes of the lines are so different. Identical slopes would indicate a similar evolution of surface steps, which have been found to be so important for the nucleation and growth of high-T_c films. Thus, we note that the rare-earth oxides may be suitable for trilayer-type structures, but not for multilayered structures.

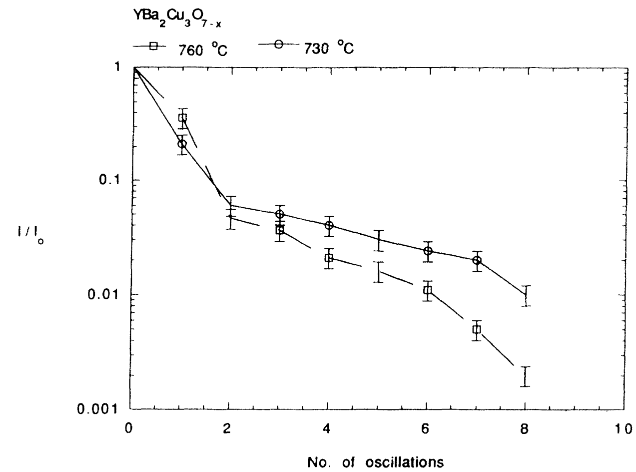

FIG. 8. Normalized oscillation intensities for $YBa_2Cu_3O_{7-x}$ films grown at different temperatures. The behavior is different from that seen in Fig. 7.

In Fig. 10, we present a planar view transmission-electron-microscope (TEM) micrograph of a $DyBa_2Cu_3O_{7-x}$ film on a $(100)SrTiO_3$ substrate. The film is nominally 100 Å thick, as evaluated by the deposition time and the cation fluxes that were used for the growth. 12 periods of RHEED intensity oscillations were monitored. Bare areas of the substrate, as evidenced by an absence of Moire fringe contrast are clearly visible. An elementary calculation of the diffracted intensity from a one-unit-cell-thick film indicates that a 5% incident intensity contribution would exist for the 103 film reflection. A modern electron-diffraction apparatus is

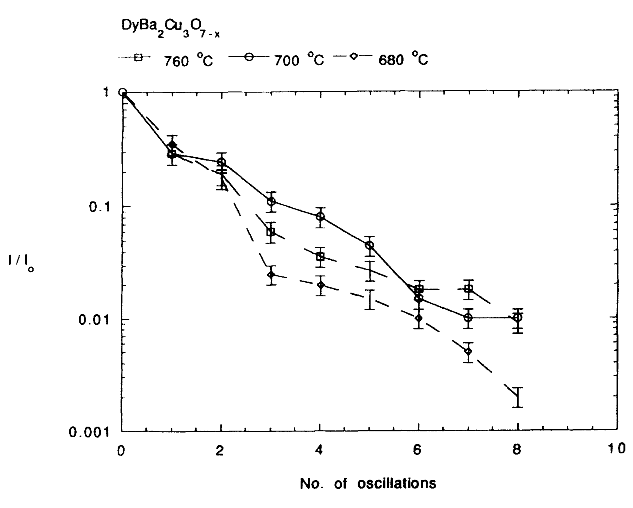

FIG. 7. Normalized oscillation intensities for $DyBa_2Cu_3O_{7-x}$ films grown at different temperatures. Reasonable coincidence is observed.

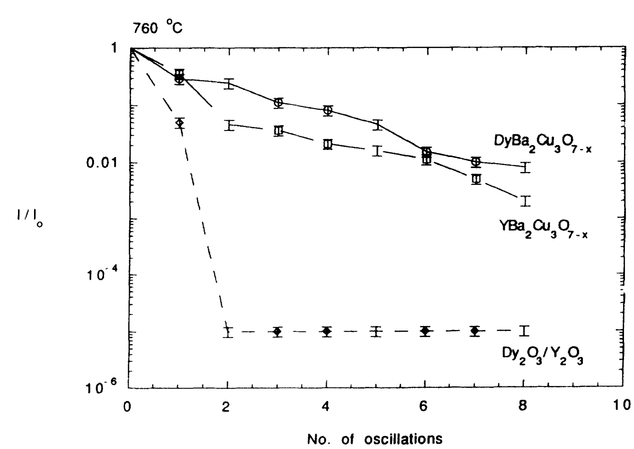

FIG. 9. Comparison of intensity oscillation data on rare-earth oxides and superconductors. The differences in slopes is suggestive of the incompatability of rare-earth oxides for multilayered structures. This is due to the differences in the evolution of the surface steps.

FIG. 10. Bright field TEM micrograph of a nominally 100-Å-thick film of $DyBa_2Cu_3O_{7-x}$ on (100)$SrTiO_3$. Bare substrate areas, as evidenced by lack of Moire fringe contrast, are clearly visible.

capable of detecting a 3% intensity contribution and we observed no such contribution from the areas where Moire fringes are absent.

The spiral morphology of grains that has been observed by various workers[16,17] is also supportive of a step edge model and our interpretation of the origin of RHEED intensity oscillations. A spiral is a natural consequence of step edge growth and a layer-by-layer growth model affords no physical explanation for their occurrence. In a previous publication, we have reported the observation of spiral growth morphology and RHEED oscillations on the same film, suggesting that atomic level smoothness is not a requirement for RHEED oscillations in these systems.[18] In contrast, the Lent and Cohen[5] picture for the origin of RHEED intensity oscillations requires atomic level smoothness and is clearly inappropriate for this system. The rounded peaks indicate a dynamical origin, as against the Lent and Cohen picture,[5] which is a kinematic approach, and is incapable of explaining the strong damping which is a dominant feature of RHEED oscillations observed in these systems. Assertions of superconductivity in single unit-cell-thick layers of these materials, where RHEED has been used to control growth, with one intensity oscillation as a signature of the completion of one unit cell, are clearly open to question.[4]

VI. CONCLUSIONS

RHEED intensity oscillations during the growth of $YBa_2Cu_3O_{7-x}$ and $DyBa_2Cu_3O_{7-x}$ on (100)$SrTiO_3$ substrates have been investigated. These studies indicate that an interpretation of the intensity oscillations in terms of changing surface step densities is appropriate. We find no basis for a layer-by-layer growth model in these systems. Experiment and theory are found to be consistent and in excellent qualitative agreement. These results are also found to be consistent with studies of nucleation and growth of these materials conducted by imaging techniques such as TEM and scanning tunneling microscope (STM). A relation between film thickness and the number of RHEED oscillations observed, was found to be nonexistent. Our study indicates that claims of superconductivity in single unit-cell-thick layers of these materials are open to question.

ACKNOWLEDGMENTS

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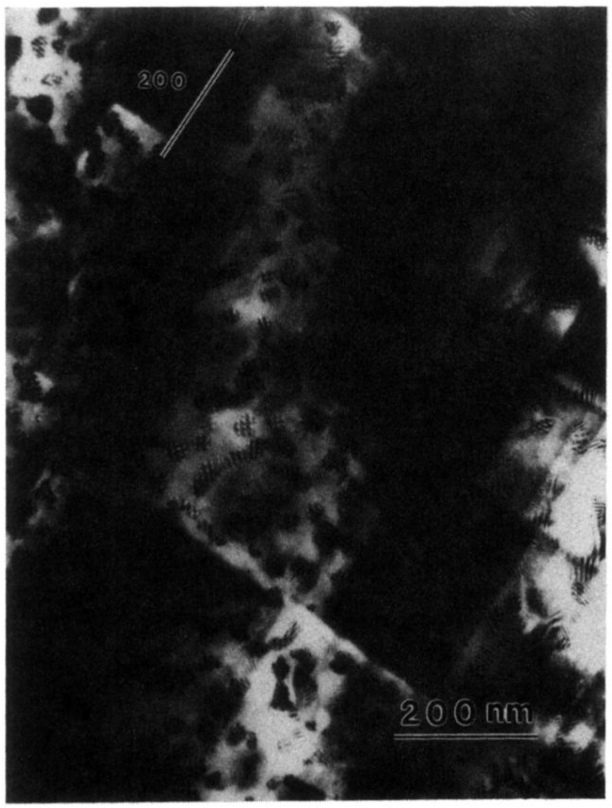

FIG. 10. Bright field TEM micrograph of a nominally 100-Å-thick film of $DyBa_2Cu_3O_{7-x}$ on (100)$SrTiO_3$. Bare substrate areas, as evidenced by lack of Moire fringe contrast, are clearly visible.

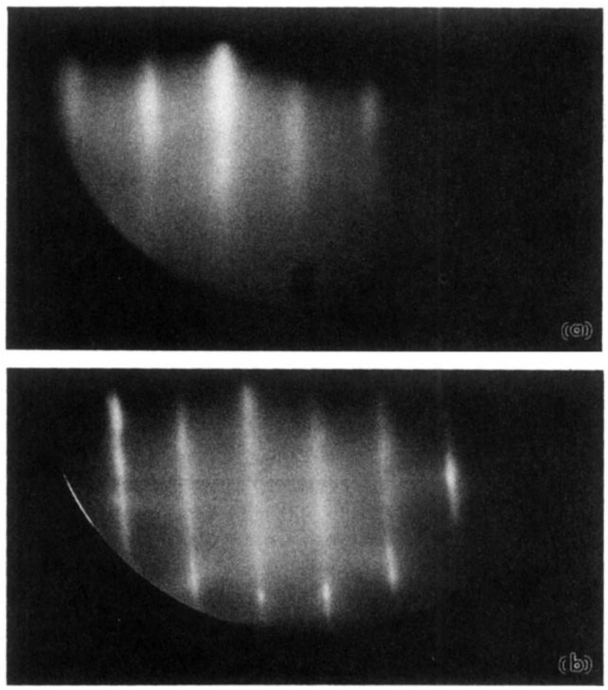

FIG. 3. RHEED patterns during the growth of a $DyBa_2Cu_3O_{7-x}$ film on (a) normal and (b) vicinal 2.2° off (100)$SrTiO_3$ substrates. The finer streaks in (b) are indicative of better film morphology.