Epitaxial SrTiO₃ film on silicon with narrow rocking curve despite huge defect density

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The structural perfection and defect microstructure of epitaxial (001) SrTiO₃ films grown on (001) Si was assessed by a combination of x-ray diffraction and scanning transmission electron microscopy. Conditions were identified that yield 002 SrTiO₃ rocking curves with full width at half-maximum below 0.03° for films ranging from 2 to 300 nm thick, but this is because this particular peak is insensitive to the $\sim 8 \times 10^{11}$ cm⁻² density of threading dislocations with pure edge character and extended defects containing dislocations and out-of-phase boundaries. Our results show that one narrow rocking curve peak is insufficient to characterize the structural perfection of epitaxial films.

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The integration of functional oxides with the backbone of semiconductor technology, silicon, has the potential to make the exceptional functional properties of oxides and oxide interfaces available in mainstream semiconductor devices. Many functional oxides have perovskite structure, and the ability to epitaxially integrate SrTiO₃, one of the most widely used substrates for the epitaxial growth of functional oxides, directly on (001) Si [1] provides a gateway to the functionalities of perovskite oxides on silicon [2]. Though SrTiO₃ turned out to be unsuitable as a high-dielectric-constant alternative gate oxide on silicon due to its small conduction-band offset [3], SrTiO₃-templated silicon offers a sound platform for integrating the unique properties of functional oxides with silicon.

Examples of the phenomena and functional properties that have been achieved on SrTiO₃-buffered silicon are as follows: ferroelectricity utilizing Pb(Zr, Ti)O₃ [4,5] and ultrathin SrTiO₃ [6], ferromagnetism utilizing La_{0.7}Sr_{0.3}MnO₃ [7,8], multiferroicity utilizing BiFeO₃ [9], piezoelectricity utilizing Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ (PMN-PT) [10], flexoelectricity utilizing SrTiO₃ [11], superconductivity utilizing YbBa₂Cu₃O_{7-x} [12], electro-optical properties utilizing BaTiO₃ [13], two-dimensional electron gases utilizing the LaAlO₃/SrTiO₃ interface [14] and the LaTiO₃/SrTiO₃ interface [15], and the photocatalysis of water reduction utilizing a thin epitaxial SrTiO₃ protection layer [16]. Thanks to the welldeveloped Czochralski method of growing large-scale silicon single crystals, growing SrTiO₃ on silicon also enhances scalability to large diameter substrates [17,18]. This is in contrast with the growth on $SrTiO_3$ single-crystal substrates, which are currently restricted by the diameter of high-quality $SrTiO_3$ at or below 2 in. [19]. The ability of epitaxial $SrTiO_3$ -on-silicon to facilitate the jump to large substrates—provided the $SrTiO_3$ layer has sufficient structural perfection—could be game-changing for oxide electronics, paving a way to industrialize multifunctional thin films for devices.

Various techniques have been employed in growing $SrTiO_3$ films on silicon, including thermal evaporation [1], molecularbeam epitaxy (MBE) [20–24], and pulsed-laser deposition (PLD) [25]. Among these techniques, MBE is the most reliable for growing epitaxial $SrTiO_3$ films on silicon [26–28], thanks to its ultrahigh-vacuum environment, exquisite control of atomic layering, and gentle growth conditions (the kinetic energies of the supplied species are all <1 eV).

A key step in the growth of epitaxial SrTiO₃ thin films on silicon is to deposit a submonolayer of strontium metal on the silicon substrate prior to the growth of SrTiO₃ [1,20,21,24]. The optimal dosage on a clean (001) Si surface is a half a monolayer [29]. It is widely recognized that this submonolayer of strontium metal is essential to the epitaxial growth of $SrTiO_3$ on silicon [20,24,27,29,30], and it may play a role in the phase-separation instability of 1-nm-thick SrTiO₃ on silicon [31]. An important question, which has not been systematically studied, is the influence of the deposition temperature of this half a monolayer of strontium on the growth of SrTiO₃ films on silicon, especially with regard to the crystalline quality of the SrTiO₃ film. It has been shown that the deposition temperature of the submonolayer of strontium metal can influence the crystalline quality of epitaxial BaO films grown on silicon [32], but there are no quantitative investigations of the influence of the deposition temperature

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of half a monolayer of strontium on the crystalline perfection of $SrTiO_3$ films on silicon.

In this paper, we study the effect of the deposition temperature of the initial half-monolayer of strontium metal on silicon on the x-ray diffraction (XRD) rocking curves (ω scans) of the 002 SrTiO₃ peaks. Narrow XRD rocking curves are usually considered synonymous with high structural quality [33,34]. Our results indicate that when the half a monolayer of strontium is deposited at substrate temperatures in the 200-800 °C range, SrTiO₃ films on silicon with a rocking curve full width at half-maximum (FWHM) of the 002 SrTiO₃ peak comparable to that of typical $SrTiO_3$ single crystals [35–38] can be achieved for films over the entire range of film thickness studied (2-300 nm). Despite the record narrow FWHM of the 002 SrTiO₃ rocking curve, measuring XRD ϕ scans of the 101 SrTiO₃ peaks in the same films reveals that significantly greater disorder is found to exist in-plane than out-of-plane. To characterize the defect microstructure in these SrTiO₃ films in greater detail, atomic resolution scanning transmission electron microscopy (STEM) reveals a threading dislocation density about six orders of magnitude higher than typical SrTiO₃ single crystals [39–41]. Thus, we conclude that a single narrow rocking curve peak is insufficient to characterize the structural perfection of epitaxial films.

All SrTiO₃ films in this study were grown in a Veeco GEN10 system on commercial 3-in.-diam (001) Si wafers. Elemental strontium and titanium beams were evaporated from a conventional low-temperature effusion cell and a Ti-Ball [42], respectively. Molecular oxygen was introduced and controlled via a piezoelectric leak valve. The temperature of the silicon substrate was monitored with either an optical pyrometer with a measurement wavelength of 980 nm (for substrate temperatures above 500 °C) or a thermocouple (for substrate temperatures below 500 °C). In situ reflection highenergy electron diffraction (RHEED) was used to monitor the growth. The crystalline perfection of the SrTiO₃ films was assessed *ex situ* by XRD θ -2 θ , rocking curve (ω scans), and ϕ scans, using either a Rigaku SmartLab or a PANalytical X'pert system with Cu $K\alpha_1$ radiation. The film microstructure was characterized by STEM using an aberration corrected FEI Titan Themis 300 operated at 300 keV.

Prior to the growth of each SrTiO₃ film, the strontium and titanium fluxes were precisely matched at $\sim 10^{13}$ atoms cm⁻² s⁻¹ via a combined shuttered and codeposition RHEED calibration method [6,43,44]. The silicon substrate was cleaned in an ultraviolet ozone cleaner for ~ 20 min to remove organic contamination, before being loaded into the MBE growth chamber with a base pressure in the high 10^{-9} Torr range. The native surface SiO₂ of the silicon substrate was removed by heating the silicon substrate to \sim 980 °C in ultrahigh vacuum for \sim 20 min. Next, the clean silicon surface was dosed with a half of a monolayer of strontium $(3.4 \times 10^{14} \text{ atoms cm}^{-2})$ at substrate temperatures ranging from ~200 to ~850 °C; at high temperature, not all of the incident strontium remained on the substrate surface. Finally, the substrate was cooled down to 200-300 °C for the growth of the SrTiO₃ film. This is the most challenging part of the growth process [21].

One of the difficulties of growing SrTiO₃ films on silicon is the initiation of the SrTiO₃ growth, where oxygen should be

introduced into the chamber to oxidize strontium and titanium atoms to form SrTiO₃, yet the silicon surface should be protected from oxygen to avoid forming an amorphous SiO₂ layer that precludes epitaxial growth. Several methodologies have been described to initiate the growth of SrTiO₃ on silicon. The crystalline qualities of the resulting SrTiO₃ films on silicon vary significantly from method to method. In one method, after depositing the submonolayer strontium, it is exposed to oxygen prior to the deposition of the SrTiO₃ film [45-47]. In another method, SrO and TiO₂ layers are deposited at a sufficiently low substrate temperature that they are initially amorphous, and then during a subsequent vacuum annealing step they are recrystallized into SrTiO₃ [15,24,26]. For the method we use in this study, the SrTiO₃ film is grown by codepositing (strontium, titanium, and oxygen are all supplied simultaneously) 2.5 unit cells of SrTiO₃ at a temperature below 300 °C (but sufficiently warm that the deposited layers remain crystalline) without previously exposing the submonolayer strontium to oxygen. While the principle of the growth follows the epitaxy-by-periodic-annealing method [21], great care is taken at the stage when oxygen is introduced into the vacuum chamber.

To be specific, the strontium and titanium shutters are kept closed as the oxygen is first introduced and the background oxygen pressure ramps up. When the oxygen partial pressure reaches $\sim 5 \times 10^{-9}$ Torr, the strontium and titanium shutters are opened simultaneously as the oxygen partial pressure slowly makes its way to $(5-7) \times 10^{-8}$ Torr. The proper ramping rate of this oxygen initiation process is key for growing SrTiO₃ films of relatively high crystalline quality on silicon [21,48]. If the strontium and titanium shutters are opened too early before the oxygen partial pressure reaches $\sim 5 \times 10^{-9}$ Torr, or the shutters are opened too late when a considerable amount of oxygen (with oxygen partial pressure much larger than 5×10^{-9} Torr) is already established in the chamber, the crystalline quality of the SrTiO₃ film is severely degraded. Following the growth of the 2.5-unitcell-thick SrTiO₃ layer, the oxygen valve is closed and the remaining oxygen is pumped out of the chamber to an oxygen partial pressure below $\sim 1 \times 10^{-9}$ Torr. The 2.5-unit-cell-thick SrTiO₃ film is then annealed at \sim 580 °C for \sim 8 min in vacuum to enhance its crystalline quality, before being cooled down to \sim 300 °C for the deposition of an additional 2.5-unitcell-thick SrTiO₃ layer. For the growth of the second 2.5-unitcell-thick SrTiO₃ layer, the strontium and titanium shutters do not necessarily need to be opened immediately after the oxygen partial pressure reaches 5×10^{-9} Torr because the first 2.5 unit-cells of SrTiO₃ serve as an oxygen diffusion barrier and help to protect the silicon substrate from being oxidized.

A schematic of the growth parameters used for the first and the second 2.5-unit-cell-thick SrTiO₃ layers, as well as one set of actual growth parameters (substrate temperature and oxygen partial pressure), are shown in Figs. 1(a) and 1(b), respectively. This epitaxy-by-periodic-annealing method was repeated three times to achieve a total SrTiO₃ film thickness of 7.5 unit cells. Upon this SrTiO₃ buffer layer, additional SrTiO₃ was grown by codeposition at a temperature of ~580 °C and an oxygen partial pressure of ~(5 – 7) × 10^{-8} Torr.



FIG. 1. (a) Schematic drawing of the growth pathway for the epitaxy-by-periodic-annealing method of the initial 5 unit-cells of SrTiO₃ on silicon. T_{sub} is the substrate temperature and P_{O_2} is the oxygen partial pressure. Different colors indicate different stages. Note the difference of the O₂ introduction (O₂ input) step between the first 2.5 unit-cells and the second 2.5 unit-cells of SrTiO₃ growth. (b) A set of actual growth parameters recorded during the growth of 5 unit-cells of SrTiO₃ on silicon. *P* is the background oxygen partial pressure measured by the chamber ion gauge. T_{sub} is the substrate thermocouple temperature.

SrTiO₃ films on silicon with total thickness ranging from 2 to 300 nm were grown using the meticulously controlled growth pathway described above. Figure 2 shows RHEED images at different growth stages from a 20-nm-thick SrTiO₃ film. These RHEED images are from the same sample for which the growth parameters are shown in Fig. 1(b). No other phases were detected in the RHEED patterns; the 20-nm-thick SrTiO₃ film was epitaxial and single-phase. The sharp and streaky RHEED patterns indicate that the surface of the SrTiO₃ film is smooth.

Figure 3(a) shows the XRD θ -2 θ scan of the same 20-nmthick SrTiO₃ film on silicon. The presence of only *00l* Bragg reflections in combination with the RHEED patterns in Fig. 2 show that the as-grown $SrTiO_3$ film is epitaxial. Figure 3(b) shows the corresponding rocking curve of the 002 SrTiO₃ peak. The narrow rocking curve implies that the SrTiO₃ film on silicon possesses a low degree of mosaic spread along the out-of-plane direction. Note, as we discuss in more detail below, that the observation of a narrow 002 rocking curve peak only implies that the density of threading dislocations that are screw dislocations with an out-of-plane line direction (or have a screw component with an out-of-plane line direction) in the SrTiO₃ film is low. The film could have high densities of threading dislocations that are pure edge dislocations with an out-of-plane line direction as such dislocations will not cause broadening in the out-of-plane direction; this has been shown nicely in (0001) GaN films grown on (0001) Al₂O₃ substrates,



FIG. 2. RHEED images at different stages during the growth of the 20-nm-thick $SrTiO_3$ film on silicon. A half a monolayer of strontium was deposited at ~800 °C, followed by another half a monolayer of strontium deposited at ~300 °C. (a) The as-grown and (c) annealed first 2.5 unit-cells of $SrTiO_3$ viewed along the [100] azimuth of (001) $SrTiO_3$. (b) The as-grown and (d) annealed first 2.5 unit-cells of $SrTiO_3$ viewed along the [110] azimuth of (001) $SrTiO_3$. RHEED patterns after the growth of the 20-nm-thick $SrTiO_3$ film viewed along (e) the [100] azimuth and (f) the [110] azimuth of (001) $SrTiO_3$.

where the rocking curve FWHM of the 0002 GaN peak was found to be 0.011° despite the presence of $\sim 2 \times 10^{10}$ cm⁻² threading edge dislocations [49].

With a FWHM of only $\sim 0.0054^{\circ}$, the rocking curve in Fig. 3(b) is the narrowest 002 SrTiO₃ rocking curve among all SrTiO₃ films on silicon reported in the literature [27], including postannealed SrTiO₃ films on silicon [50,51]. It is even narrower than the 002 rocking curve of typical SrTiO₃ single crystals [35-38]. Figures 3(c) and 3(d) show a two-dimensional rocking curve of a 10-nm-thick SrTiO₃ film on silicon and the corresponding reciprocal space map (RSM) around the 002 SrTiO₃ peak, respectively. The narrow spreading of the RSM is consistent with the rocking curve measurement, corroborating the high crystalline perfection of the SrTiO₃ film along the out-of-plane direction. In addition to the lab XRD measurement, we confirmed the peak broadening of the same 10-nm-thick SrTiO₃ film on silicon with synchrotron diffraction measurements. The RSM of the same sample measured at the Cornell High Energy Synchrotron Source (CHESS) agrees with the lab XRD measurement, as Fig. 3(e) shows. With our refined growth method, SrTiO₃ films as thin as 5 unit-cells thick can be measured with a lab XRD. A θ -2 θ scan of a 5-unit-cell-thick SrTiO₃ film on silicon and a rocking curve of the 002 SrTiO₃ peak of the same film are shown in Figs. S1(a) and S1(b) in the Supplemental Material [52].

To test whether the narrow FWHM of the 002 rocking curve (a symmetric peak) is indicative that the film has a low density of threading dislocations (as would follow from classic work in which the dislocations that broaden the rocking curve are assumed to be randomly distributed) [33], the rocking curve of an asymmetric peak was also measured. Figure 3(f) shows the rocking curve of the 103 SrTiO₃ peak and the 002 SrTiO₃ peak (with intensities normalized) measured on a 300-nm-thick SrTiO₃ film on silicon. In contrast with the very narrow rocking curve of the 002 SrTiO₃ peak,



FIG. 3. XRD using (a)–(d), (f), (g) laboratory, and (e) sychrotron diffractometers of two SrTiO₃ films grown on silicon in this study. (a) θ -2 θ scan of the 20-nm-thick SrTiO₃ on silicon shows only 00l Bragg reflections, which in combination with the RHEED images indicates an epitaxial SrTiO₃ film on silicon. Asterisks indicate peaks arising from the silicon substrate. (b) Rocking curve of the 002 SrTiO₃ peak of the 20-nm-thick SrTiO₃ on silicon has a FWHM of ~0.0054°. (c) Rocking curves around the 002 SrTiO₃ peak of the 10-nm-thick SrTiO₃ on silicon, along with (d) its corresponding RSM exhibit peak broadening consistent with the rocking curve. The broadening in the θ -2 θ scan (a) is caused by the finite thickness of the film, while the confinement along the ω direction shows low mosaic spread in the out-of-plane direction. For the RSM, the broadening along the Q_z direction is due to the finite thickness of the film, while the narrow width in the Q_x direction indicates the low mosaic spread of the SrTiO₃ film in this direction. (e) The RSM of the 10-nm-thick SrTiO₃ on silicon with clear thickness fringes measured at the G2 station of CHESS indicates that the narrow intensity spread along the Q_x direction and the broadening along the Q_z direction are consistent with the lab XRD measurement. For the 10-nm-thick and 20-nm-thick samples that are shown in Fig. 2 and Fig. 3, we first deposited half a monolayer of strontium at high temperature (>= 700 °C), and then deposited another half a monolayer of strontium at 300 °C. Normally we only deposit half a monolayer of strontium once at a certain temperature, before the growth of the SrTiO₃ film. (f) Rocking curves of the 103 and 002 SrTiO₃ peaks of the 300-nm-thick SrTiO₃ film on silicon. (g) XRD pole figure measurements sampling the 202 Si and 101 SrTiO₃ family of peaks of the 20-nm-thick SrTiO₃ on silicon. The intensity color scale is logarithmic.

the rocking curve of the 103 SrTiO₃ peak is about eight times broader. This significant difference in the rocking curve width is consistent with the defect microstructure revealed by STEM (described at the end of the paper). Specifically, it is consistent with the film containing a high density of threading dislocations, and the narrow FWHM observed for the 002 peak is due to these threading dislocations being predominantly edge dislocations with out-of-plane line direction analogous to what was observed for some (0001) GaN films grown on (0001) Al₂O₃ substrates [49].

XRD pole figure measurements sampling the 111 Si and 101 SrTiO₃ families of peaks of the same 20-nm-thick SrTiO₃

on silicon characterized in Fig. 1 are shown in Fig. 3(g). The radial coordinate and the angular coordinate correspond to the XRD χ angle and the ϕ angle, respectively. For the entire range (0° < χ < 90°, 0° < ϕ < 360°) measured, only the 111 Si family of peaks (χ = 54.73°, 2 θ = 28.44°) and the 101 SrTiO₃ family of peaks (χ = 45°, 2 θ = 32.4°) were detected, indicating that the epitaxial relationship is (001) Si||(001) SrTiO₃, [110] Si||[100] SrTiO₃.

The influence of the deposition temperature of the initial half-monolayer of strontium on the bare silicon substrate on the crystalline quality (as judged by the FHWM of the 002 rocking curve) of the SrTiO₃ film was investigated by growing

20-nm-thick SrTiO₃ films and depositing strontium at temperatures from ~200 to ~850 °C. To ensure a half a monolayer of strontium, the silicon was thermally cleaned rather than using strontium-assisted deoxidation [53]. This is because the latter method leaves a fractional monolayer of strontium atoms on the silicon surface after the strontium-assisted deoxidation. At each strontium deposition temperature, multiple 20-nm-thick SrTiO₃ films were grown and the film with the narrowest FWHM of the rocking curve for each temperature was chosen for comparison. Figure 4(a) shows the smallest FWHM value of the 002 SrTiO₃ peak of 20-nm-thick SrTiO₃ films obtained as a function of deposition temperature, indicating that 20-nm-thick SrTiO₃ films with narrow rocking curves can be grown over a wide range of strontium deposition temperatures: ~200 to ~800 °C.

At a deposition temperature of ~850 °C, the rocking curve of the 002 SrTiO₃ peak is significantly broader. This might be due to the diffusion of strontium atoms into the bulk silicon substrate or evaporation from the surface of the substrate at such a high temperature. Either way, the half-monolayer strontium template for epitaxial SrTiO₃ growth would be incomplete. Even though the 002 rocking curve of the SrTiO₃ film is degraded when the half-monolayer of strontium is deposited at ~850 °C, a phase-pure 20-nm-thick SrTiO₃ film with a θ -2 θ scan containing only 001 Bragg reflections still results.

It was previously regarded as difficult or impossible to grow crystalline oxides on silicon when the half-monolayer of strontium was deposited at low temperatures and a silicide layer was not formed, say at ~ 200 or $\sim 300 \,^{\circ}$ C [32]. Indeed, earlier work utilized temperatures $\geq 600 \,^{\circ}$ C for the deposition of the strontium on bare silicon prior to depositing SrTiO₃ on top of it [15,20,22,23,54]. Although the RHEED patterns of the as-grown first 2.5 unit-cells of SrTiO₃ are not sharp, we find (see Fig. S2 in the Supplemental Material [52]) that when the half a monolayer of strontium is deposited at ~ 200 or \sim 300 °C, after recrystallization in vacuum at \sim 580 °C, the FWHMs of the 002 SrTiO₃ rocking curves are comparable to that of the SrTiO₃ films grown on a half-monolayer of strontium deposited at higher temperatures. The XRD θ -2 θ scan and rocking curve of a 60-nm-thick SrTiO₃ with its half a monolayer of strontium deposited at \sim 300 °C are shown in Fig. S3 in the Supplemental Material [52].

When the half-monolayer of strontium is deposited at 850, 650, or 300 °C, it induces different (001) Si surface reconstructions, which can be shown by measuring the RHEED intensity of the half-order streak along the [110] azimuth of (001) Si as a function of strontium coverage. Figure S4 in the Supplemental Material [52] shows this evolution for a half-monolayer of strontium deposited at 850, 650, and 300 °C. Although the evolution of the RHEED intensity as a function of strontium coverage changes with substrate temperature, our growth results show unambiguously that a half-monolayer of strontium can serve as a template for the growth of epitaxial SrTiO₃ films on silicon with narrow 002 SrTiO₃ rocking curves no matter whether it forms the so-called silicide layer [20] or stays (partially) physisorbed to the silicon surface [32].

Prior reports show $SrTiO_3$ films with a narrow 002 $SrTiO_3$ rocking curve on silicon for film thickness less than about 2 nm, where the $SrTiO_3$ is commensurately strained to the

underlying silicon substrate [6] or for film thicknesses above about 100 nm [17,50,55]. At intermediate thicknesses, the films show higher rocking curve FWHMs of the 002 SrTiO₃ peak. Such behavior is typical for the growth of mismatched epitaxial films [56]. Our work extends the thickness range over which films with narrow rocking curves are obtained from \sim 5 unit-cells to \sim 300 nm, as is shown in Fig. 4(b). A comparison with representative SrTiO₃ films on silicon from the literature [6,17] is included in Fig. 4(b). Raw data of the rocking curves of the 002 SrTiO₃ peak of SrTiO₃ films of different thicknesses are shown in Fig. 4(c). Judging from the narrow rocking curve of the 002 SrTiO₃ peak, we believe that the half-monolayer of strontium on silicon not only acts as a diffusion barrier for protecting the (001) Si surface from being oxidized during the initial SrTiO₃ growth, but it also influences the type of threading dislocations that subsequently form. In our case, these threading dislocations are predominantly edge dislocations with an out-of-plane line direction, which is consistent with the extremely narrow FWHM of the 002 rocking curves as well as the STEM results described below. The out-of-plane lattice parameter of the SrTiO₃ films as a function of the film thickness is shown in Fig. 4(d), from which we see that the SrTiO₃ film relaxes quickly after its thickness is above about 5 unit-cells.

Measurements of the rocking curves of the 003 and 004 SrTiO₃ peaks indicate that our SrTiO₃ films on silicon of intermediate thicknesses such as 20 nm thick can be described by the unconventional mosaic crystal model [57]. To be specific, the diffuse or the broad tail of the rocking curve comes from the short-range order of independent scattering of individual dislocations, while the sharp peak seen from the 002 SrTiO₃ peak is due to the long-range correlation of the SrTiO₃ film, and is not seen in higher-order peaks such as the 003 and 004 SrTiO₃ peaks [57]. Figure S5 in the Supplemental Material [52] shows the rocking curves of the *OOl* SrTiO₃ peaks (l = 1, 2, 3, and 4) of a 40-nm-thick SrTiO₃ film on silicon. Only the rocking curves of the 001 and 002 SrTiO₃ peaks are narrow; the same does not hold for the 003 and 004 SrTiO₃ peaks. This implies that the SrTiO₃ film on silicon is a so-called unconventional mosaic crystal [57].

We measured the azimuth rotational disorder of the SrTiO₃ films on silicon via XRD ϕ scans. As an example, a 60-nm-thick SrTiO₃ on silicon film shows a relatively large FWHM of $\sim 0.75^{\circ}$ of the 101 SrTiO₃ peak for a ϕ scan (Fig. S6 in the Supplemental Material [52]), which implies relatively large in-plane mosaic spread. The rocking curve FWHM of the 002 SrTiO₃ peak of this same 60-nm-thick film is ~0.012°. The FWHM of azimuthal ϕ scans as a function of the film thickness is shown in Fig. 4(e), indicating that the rotational disorder of the SrTiO₃ film decreases as the film thickness increases. The existence of a considerable amount of in-plane rotational disorder of SrTiO₃ films on silicon can be attributed to the lattice mismatch as well as the relatively large difference between the thermal expansion coefficient of silicon (averaging $3.5 \times 10^{-6} \text{ K}^{-1}$ between room temperature and 520 °C) [58] and that of SrTiO₃ (averaging $1.08 \times 10^{-5} \text{ K}^{-1}$ between room temperature and 520 °C) [58,59]. The large difference between thermal expansion coefficients between the substrate and the film can introduce extra misfit between the film and the substrate,



FIG. 4. (a) The rocking curve FWHMs of 20-nm-thick films of SrTiO₃ on silicon as a function of the temperature ($T_{\text{strontium deposition}}$) at which the half a monolayer of strontium was deposited show that 20-nm-thick SrTiO₃ films with narrow rocking curves can be grown when the half a monolayer of strontium is deposited anywhere in the ~200 to 800 °C range. Rocking curves of the 002 SrTiO₃ peak were measured. All data points are from 20-nm-thick SrTiO₃ films grown on silicon with θ -2 θ scans showing only 00l Bragg reflections. (b) FWHMs of SrTiO₃ films of different thicknesses on silicon show that from ~2 to ~300 nm, our growth method yields films with narrow FWHM. The data points of this work are represented with red stars. The purple data points and blue data points are from Refs. [6] and [54], respectively. (c) Raw data of the rocking curves of the 002 SrTiO₃ peak of SrTiO₃ films of different thicknesses. (d) Out-of-plane lattice parameter of the SrTiO₃ film on silicon as a function of film thickness showing that the SrTiO₃ film on silicon relaxes quickly above a film thickness of about 5 unit-cells. (e) The FWHM of the 101 SrTiO₃ peak in ϕ (measured in a triple-axis geometry) for SrTiO₃ films on silicon as a function of film thickness a relatively large in-plane rotational disorder, and the in-plane mosaicity spread decreases as the film thickness increases.



FIG. 5. (a) STEM images of a 10-nm-thick SrTiO₃ film on silicon. The interfacial region has an amorphous SiO₂ layer. Strontium (orange), titanium (blue), and silicon (red) atoms are overlaid to show the schematic atomic structure. (b) Plan-view STEM image of a 20-nm-thick SrTiO₃ film on silicon showing a variety of both localized and branching dislocations with a density of $\sim 8 \times 10^{11}$ cm⁻², between which exist crystallographically defect-free regions with area on the order of $\sim (20 \text{ nm})^2$. (c) Atomic resolution STEM image of a branching defect like those seen in (b), with an out-of-phase boundary highlighted by yellow arrows marking rows of bright strontium atoms offset on either side of the fault. (d) Atomic resolution STEM image of a localized edge dislocation like those seen in (b) with a Burgers circuit shown in yellow.

which can yield additional in-plane rotational disorder during the cool-down process after the film growth [60].

Cross-sectional high-angle annular dark field (HAADF) STEM was used to clarify the defect microstructure and characterize the interface between the SrTiO₃ and silicon. A representative image of the interface between a 10-nmthick SrTiO₃ film and silicon substrate is shown in Fig. 5(a). Although we used only the epitaxy-by-periodic-annealing method (with no high-temperature growth step) to grow the SrTiO₃ film, we see an amorphous SiO₂ layer at the interface between the SrTiO₃ film and the silicon substrate. This oxidation of the surface of the silicon either came from the diffusion of oxygen through the SrTiO₃ film during the ~300 °C deposition steps, or more likely during the vacuum annealing step at 580 °C.

We also performed plan-view STEM to characterize the microstructure of our $SrTiO_3$ on silicon films. As is clearly visible in Fig. 5(b), the films show a high density of crystalline defects. While some of the defects can be easily identified by a simple Burgers circuit, as shown in Fig. 5(d), many others have a more complicated extended and branching structure, like the one shown in Fig. 5(c). By following the rows

of strontium sites (yellow arrows) across the defect shown in Fig. 5(c), a clear out-of-phase boundary [61,62] can be seen. Interestingly, the boundary shown here—as in many of the defects observed in these films—runs along the [110] a_{SrTiO3} direction corresponding to an (a+b)/2 unit-cell shift. Toward the termination of the defect, the exact structure in the HAADF STEM image becomes less clear. Nonetheless, Fourier peak analysis suggests that the structure in this and similar regions can be well described by a combination of threading dislocations and out-of-phase boundaries, as shown in Fig. S7 in the Supplemental Material [52].

The underlying cause of the out-of-phase boundaries could be the coalescence of nuclei or the incorporation of planar defects. Because the step height of silicon does not match that of $SrTiO_3$, $SrTiO_3$ nuclei that form on different terraces of the silicon substrate will be out-of-phase with each other. When the growth fronts from such separate $SrTiO_3$ nuclei coalesce, out-of-phase boundaries can form [62]. Another possible nucleation mechanism is due to nonstoichiometry. If the $SrTiO_3$ is locally SrO-rich, an extra SrO rock-salt layer could be inserted into the $SrTiO_3$ perovskite structure forming a Ruddlesden-Popper fault [63,64]. These SrO double layers cause $\frac{a}{2}$ [110] shifts in the position of the SrTiO₃ unit cells on either side of such a crystallographic shear fault [65]. The resulting offset in coalescing SrTiO₃ growth fronts would again give rise to an out-of-phase boundary.

Employing the same Fourier peak analysis mentioned above, threading dislocations are easily identified and located throughout several plan-view STEM images to obtain a representative sampling. The SrTiO₃ films are found to contain a high density of threading dislocations, $\sim 8 \times 10^{11}$ cm⁻². The FWHM of the 002 rocking curve of the same film studied by STEM is $\sim 0.008^{\circ}$. Note that this density of threading dislocations is far higher than that found in typical SrTiO₃ single crystals, which lie in the $(3 - 10) \times 10^5$ cm⁻² range [39–41].

This massive difference in dislocation density begs the question of how it is possible for a SrTiO₃ film that contains a threading dislocation density more than six orders of magnitude higher than a SrTiO₃ single crystal to exhibit an 002 SrTiO₃ rocking curve with a FWHM (\sim 0.008°) that is considerably narrower than that of the single crystal? As shown in Fig. 5(d), the dominant Burgers vector is found to be [100] a_{SrTiO3} . The threading dislocations are thus pure edge dislocations with an out-of-plane line direction, and the extremely narrow 002 rocking curves arise from the insensitivity of the 002 rocking curve to these pure edge dislocations, as has been reported previously for (0001) GaN / (0001) Al₂O₃ films [49]. When dislocations are distributed anisotropically, as they are in these SrTiO₃/Si films, it is insufficient to take a single narrow rocking curve peak as evidence of structural perfection. Assessing multiple peaks becomes important to establish the structural distortions present in multiple directions.

In summary, by depositing a half-monolayer of strontium metal on bare (001) Si in the substrate temperature range 200–800 °C, in combination with a carefully controlled epitaxy-by-

periodic-annealing method, we have lowered the out-of-plane mosaic spread of the SrTiO₃ films on silicon. SrTiO₃ films of low out-of-plane mosaic spread with thicknesses from 5 unit-cells to \sim 300 nm were achieved. Our work establishes a well-tested methodology for consistently growing epitaxial SrTiO₃ films on silicon using MBE. This study has the potential to increase the quality of not only the SrTiO₃ films on silicon, but also that of overlying epitaxial functional layers as the SrTiO₃ layer serves as an epitaxial template for the integration of oxides with a multitude of functional properties with silicon [66–68].

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Supplementary Material for

Epitaxial SrTiO₃ Film on Silicon with Narrow Rocking Curve Despite Huge Defect Density

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FIG. S1. (a) θ -2 θ scan of a 5 unit-cell-thick SrTiO₃ film grown on silicon, measured with Rigaku SmartLab. The asterisk indicates the substrate peak. (b) The rocking curve (measured with PANalytical X'pert system) FWHM of the 002 SrTiO₃ peak is ~0.0044° and is comparable to that of the silicon 004 peak. The scans are offset from each other along the vertical axis for clarity.



FIG. S2. RHEED images at different stages during the growth of the 60 nm thick $SrTiO_3$ film on silicon with the half a monolayer of strontium deposited at 300 °C. (a) The bare silicon surface along the [110] azimuth of (001) Si after the deposition of a half a monolayer of strontium at ~300 °C. (b) The asgrown and (e) the annealed first 2.5 unit-cell-thick $SrTiO_3$ along the [100] azimuth of (001) $SrTiO_3$. (c) The as-grown and (f) the annealed first 2.5 unit-cell-thick $SrTiO_3$ along the [110] azimuth of (001) $SrTiO_3$. The 60 nm thick $SrTiO_3$ after growth viewed along (d) the [100] azimuth and (g) [110] azimuth of (001) $SrTiO_3$.



FIG. S3. XRD of a 60 nm thick SrTiO₃ on silicon grown with the half a monolayer of strontium metal deposited at 300 °C. (a) θ -2 θ scan and (b) the rocking curve of the 002 SrTiO₃ peak with a FWHM of ~0.017°.



FIG. S4. Comparison of the RHEED intensity change during the deposition of the half a monolayer of strontium at ~850, ~650, and ~300 °C. The intensity of the half-order streak along the [110] azimuth of (001) Si was monitored. The RHEED patterns viewed along the [110] azimuth of (001) Si after the deposition of the half a monolayer of strontium at each temperature are shown to the right. The rectangular boxes on the RHEED patterns indicate the streak that was monitored during the deposition of the half a monolayer of strontium at each temperature.



FIG. S5. Rocking curves of the 001, 002, 003, and 004 $SrTiO_3$ peaks of a 40 nm thick $SrTiO_3$ film on silicon. The narrow peak of the rocking curve is only seen for the 001 and 002 $SrTiO_3$ peaks.



FIG. S6. ϕ scan of a 60 nm thick SrTiO₃ film on silicon. The FWHM of the ϕ scan of the 101 SrTiO₃ peak is ~0.75°. The scans are offset from each other along the vertical axis for clarity.



FIG. S7. Example of Fourier peak analysis used to separate the threading dislocation and out-of-phase boundary components that make up the extended defects seen throughout the $SrTiO_3$ on silicon films. (a) The same defect ensemble shown in Fig. 5(c). (b) Filtering the orthogonal [100] peaks in the FFT (fast Fourier transform) of image (a) separates and highlights the lattice structure in both directions, making it simpler to identify lattice variations even in regions where the original real space image is quite complicated. (c) Filtering in one direction reveals a threading dislocation near one kink of the defect. (d) Filtering in the orthogonal direction highlights the out-of-phase boundary.