

硅基 BaTiO₃ 外延薄膜的晶格弛豫行为与畴结构调控

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摘 要: 钛酸钡(BaTiO₃, BTO)薄膜因其优异的电光效应在集成光子器件中具有重要的应用潜力。然而, 在硅衬底上实现高质量 BTO 薄膜的外延生长并有效调控其面内极化以实现电光系数最大化仍面临关键挑战。采用氧化物分子束外延技术, 结合原位反射高能电子衍射(RHEED)实时监控, Si 基片上的外延钛酸锶(SrTiO₃, STO)缓冲层在 12.5 个单胞层(ML)后完成晶格弛豫, 显著降低了 BTO 薄膜与 Si 衬底的晶格失配度, 为 Si 基片上外延 BTO 提供了良好的过渡层。在 STO 上外延更大晶格常数的 BTO 薄膜时, 晶格在界面至 30 nm 之间完成弛豫过程。降温时, 由于硅衬底的热膨胀系数较低, BTO 薄膜受衬底约束, 保持了弛豫后的大面内晶格常数, 从而获得了面内铁电极化。基于此方法制备的 2 in 的 300 nm BTO 晶圆膜展现出优异的晶体质量, 摇摆曲线半峰宽为 0.45°, 表面粗糙度小于 0.2 nm, $c/a = 0.994 < 1$, 并具有显著的面内多畴结构。上述原位监控技术为分析 BTO 薄膜外延过程中的晶格弛豫及畴结构调控提供了关键信息, 为硅基 BTO 电光器件集成奠定了材料基础。

关键词: 钛酸钡; 分子束外延; 硅基异质集成; 应变调控; 面内极化

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硅光子学在高性能通信与计算系统中至关重要^[1-3], 而高性能电光调制器是其核心元件。钛酸钡(BaTiO₃, BTO)因其超高的电光系数($r_{42} \approx 923 \text{ pm/V}$)^[4], 以及和硅基直接外延集成的潜力^[5-7], 成为实现紧凑、低功耗光子集成电路的理想材料^[8]。然而, 实现高性能 BTO 器件的关键瓶颈在于制备高质量且铁电畴结构可控的单晶薄膜。其中, BTO 与 Si 的高质量集成面临界面形成 SiO₂ 非晶层和晶格失配的问题。BTO 生长需要的高温高氧压环境极易导致硅表面形成非晶 SiO₂ 层, 阻碍了 BTO 与硅之间的晶格匹配和有效外延^[9], 进而影响器件的整体性能。同时, 由于 BTO($a = b = 3.992 \text{ \AA}$, $c = 4.036 \text{ \AA}$)^[10] 与 Si($5.431 \text{ \AA}$)^[11] 的晶格常数存在显著差异, 通过建立 45° 面内旋转的外延匹配关系^[12]后仍存在约 $4.0\%[(\sqrt{2} \times a_{\text{BTO}} - a_{\text{Si}}) / a_{\text{Si}} \times 100\% = 4.0\%]$ 的晶格失配, 如此大的失配容易导致高密度位错和应变弛豫,

会严重影响薄膜质量; 而采用 STO($3.905 \text{ \AA}$)^[11] 作为缓冲层可以将晶格失配降低至 1.7% 以有效缓解界面晶格失配应力并作为外延模板。McKee 等^[13]于 1998 年首次提出采用具有高真空特点的分子束外延(MBE)技术, 通过一种分步转变策略, 避免了硅表面非晶 SiO₂ 层的形成, 实现了高质量 STO 薄膜的硅基外延。基于 McKee 的这一开创性工作, 后续研究者利用 STO 作为中间缓冲层来实现 BTO 与硅的集成^[14-16]。

BTO 电光效应强烈依赖于其铁电畴结构和晶体取向。研究表明, 面内极化取向的单晶薄膜可实现电光系数最大化, 而面外取向时横向电光效应显著衰减^[17-19]。由于畴结构受生长热力学与动力学条件的显著影响, 可通过引入更大晶格常数的中间层^[7, 17, 20]提供拉伸应变或精细调控生长条件(生长温度^[21]、生长气压^[22]、退火处理^[23]等)获得

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不同取向的薄膜，但对畴结构的分析多基于事后表征^[24]，缺乏对畴结构演化过程的实时动态监测，难以揭示畴取向转变的临界条件和物理机制。

针对上述问题，本工作通过优化 MBE 工艺参数和原位 RHEED 监测技术，实现了 2in 晶圆级高质量 BTO 薄膜的可控生长，并系统地探讨了不同厚度 STO 缓冲层与 BTO 层在生长过程中的晶格弛豫行为。通过界面工程与热应变调控，实现了 STO 层应变状态对 BTO 薄膜质量和极化取向的调控。特别是通过对 STO 缓冲层与 BTO 层晶格常数演化的定量分析，确定了促进 BTO 面内极化取向形成的临界厚度，并获得了具有面内极化取向的高质量 BTO 薄膜，为实现高性能电光调制器提供了材料基础。

1 实验

1.1 样品制备

采用氢氟酸(HF, 质量分数 10%)溶液对直径 2in 的(001)硅基片进行腐蚀移除表面 SiO₂ 非晶层，随后将清洗后的样品快速转移至超高真空腔室(背景气压 $\leq 5 \times 10^{-8}$ Pa)。SrTiO₃ 的外延生长分为 2 个阶段：第 1 阶段在超高真空环境(背景气压 $< 5 \times 10^{-7}$ Pa)中，利用 MBE 技术在 200 °C 衬底温度下沉积 2.5 个单胞层的 Sr/Ti 金属，同时精确调控氧分压至 $< 5 \times 10^{-6}$ Pa；在第 2 阶段的高温退火过程中，对沉积的 2.5 个单胞层 STO 进行 500 °C 原位退火处理 8 min，同时维持超高真空环境(真空度 $< 1 \times 10^{-6}$ Pa)。重复 3 个周期后，更多的 STO 层在热偶温度 600 °C 及 6×10^{-5} Pa 的氧分压下采用共沉积方法外延生长。BaTiO₃ 层在热偶

温度 700 °C 及 6×10^{-5} Pa 氧分压下连续共沉积生长，最终获得 310 nm BTO/5 nm STO/Si 异质结构。

1.2 样品表征

用 Bruker D8 Discover 型高分辨四圆 X 射线衍射仪 $2\theta-\omega$ 扫描、摇摆曲线扫描表征薄膜的晶格结构。测试条件：Cu 靶，管电压 40 kV，管电流 40 mA。通过光谱型椭圆偏振仪(J.A. Woollam RC2)对薄膜的光学常数及厚度进行表征。用 MPF-3D Origin+型原子力显微镜对样品表面形貌进行表征以及读写铁电畴。

2 结果与讨论

2.1 硅基钛酸锶过渡层及钛酸钡薄膜的外延生长

尽管 STO 缓冲层能够有效缓解 BTO 与 Si 之间的晶格失配(从 4.0% 降至约 1.7%)，但 STO 与 Si 之间仍需构建结构匹配的界面模板层实现高质量外延生长。STO 在(001)Si 衬底上的外延生长机制如图 1 所示。首先通过一个升温过程，当温度达到 500 °C 以上，Si 表面自发形成长程有序的二聚体阵列，如图 1c 所示。该重构过程通过反射高能电子衍射(RHEED)实时监控，在 RHEED 衍射图中，沿[110]方向观察到清晰的(2×1)重构条纹，表明表面原子已重新排列成有序的二聚体结构。在此模板基础上，采用 MBE 技术精确沉积 1/2 单层(ML)覆盖度的 Sr 原子，这一特定覆盖度下能形成稳定的界面结构^[25]，这种保持特定重构的 Sr/Si 界面结构为后续 STO 薄膜的外延生长提供了结构匹配的模板层，促进了高质量 STO 薄膜的形成。

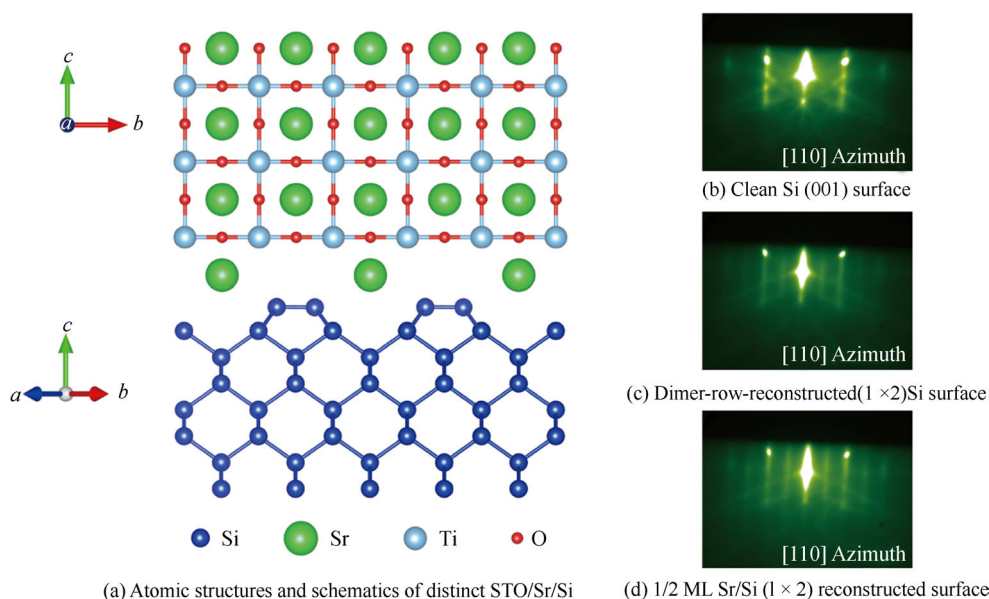
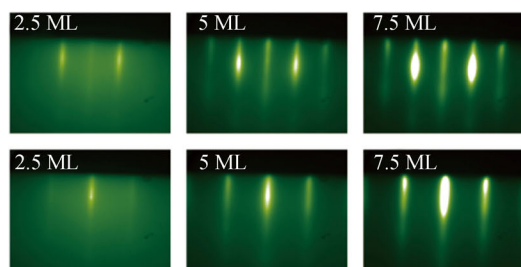


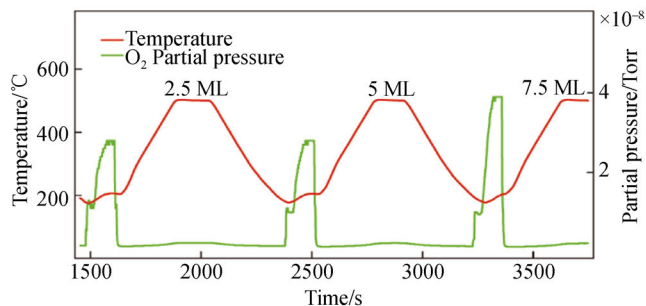
图 1 SrTiO₃ 在 Si 衬底上的外延生长机制
Fig. 1 Epitaxial growth mechanism of SrTiO₃ on Si substrate

为制备硅基高质量 STO 薄膜, 本工作采用分段外延策略, 在低温有氧沉积后再高温高真空结晶^[26-27]。图 2 为 STO 和 BTO 薄膜外延生长过程中的 RHEED 衍射图谱、光学参数及表面形貌。由图 2a~图 2b 可见, 首先是低温沉积阶段, 在氧气氛下在 Si 表面共沉积 Sr/Ti 金属, 形成非晶态 Sr-Ti-O 过渡层。沉积完成后进入高温退火阶段, 在超高真空环境下, 非晶层通过固态外延重结晶转化为单晶 STO, Si 的晶格模板诱导 STO 沿[001]方向外延。由 RHEED 衍射谱可以看出, 高温退火后, 薄膜表面出现清晰的条纹状图样条纹, 且无额外的衍射斑点, 表明未出现多晶或取向不一致的现象。随后在 5 nm STO/Si 基板上, 采用 MBE 技术连续共沉积 300 nm 厚度的 BTO 薄膜, 由图 2c 可以看出, 清晰锐利的条纹状衍射图案与菊池线表明表面原子排

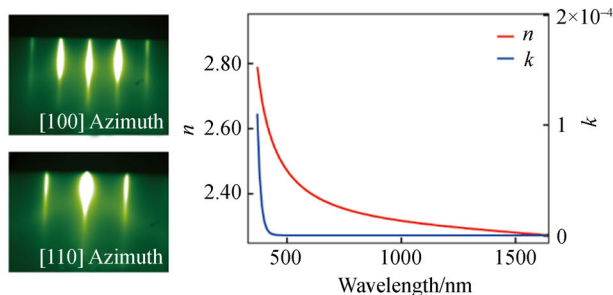
列高度有序且具有良好的晶格周期性, 证实形成了高质量的 BTO 单晶薄膜。在通信波长 1550 nm 处, BTO 薄膜呈现出理想的光学性能, 其复折射率实部 $n = 2.28$, 消光系数 k 低至 10^{-6} 量级(图 2d), 表明薄膜具有极低的光学损耗及理想的光学透明性, 为实现低损耗光电调制器奠定了材料基础。由图 2e 可见, AFM 测量显示薄膜表面达到原子级平整度, 均方根粗糙度(RMS) < 0.2 nm, 接近单晶衬底表面的天然粗糙度水平, 原子级平整表面可将器件的散射损耗降至最低。同时, 椭圆偏测量显示薄膜厚度为 (309.6 ± 2.0) nm, 与生长目标值 310 nm 的偏差仅为 0.13%, 展现了 MBE 技术在大面积薄膜制备中优异的厚度控制精度。且优异的厚度均匀性能够确保光波导模式的一致性, 是实现晶圆级器件批量制造的关键技术指标。



(a) RHEED patterns obtained along [100] azimuth (top row) and [110] azimuth (bottom row) for different film thicknesses

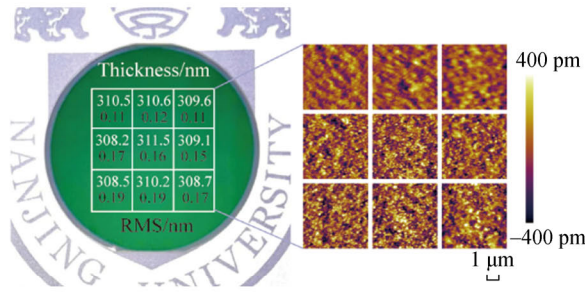


(b) Real-time growth parameters monitored during STO film deposition on silicon substrate



(c) RHEED images of 300 nm BTO

(d) Optical constants (n, k) vs wavelength for 300 nm BTO film



(e) Film thickness, roughness, and AFM topography images of 2 BTO on Si

图 2 STO 和 BTO 薄膜外延生长过程中的 RHEED 衍射谱、光学参数及表面形貌表征

Fig. 2 RHEED diffraction patterns, optical parameters, and surface morphology characterization during epitaxial growth process of STO and BTO thin films

2.2 钛酸锶过渡层与钛酸钡薄膜生长过程中的应变弛豫分析

图 3 为 STO 与 BTO 薄膜的 RHEED 衍射强度及面内晶格常数随薄膜厚度的演化过程。RHEED 衍射峰源于表面二维晶格倒易杆与 Ewald 球的交点, 其峰位间距直接反映了倒易晶格矢量的信息, 可用于分析薄膜的面内晶格常数。由图 3a 左侧

RHEED 图可见, 选取沿衬底 Si 的[110]方向入射电子束获得 RHEED 谱, 对谱中虚线标示区域的强度进行纵向积分, 以横向像素为横轴, 并以室温下 Si 衬底的晶格常数与衍射条纹间距对衍射峰位进行校准, 建立了归一化衍射强度与倒易空间矢量的关系曲线(见图 3a 和图 3c), 以及晶格常数随薄膜厚度增长的演化趋势(见图 3b 和图 3d)。

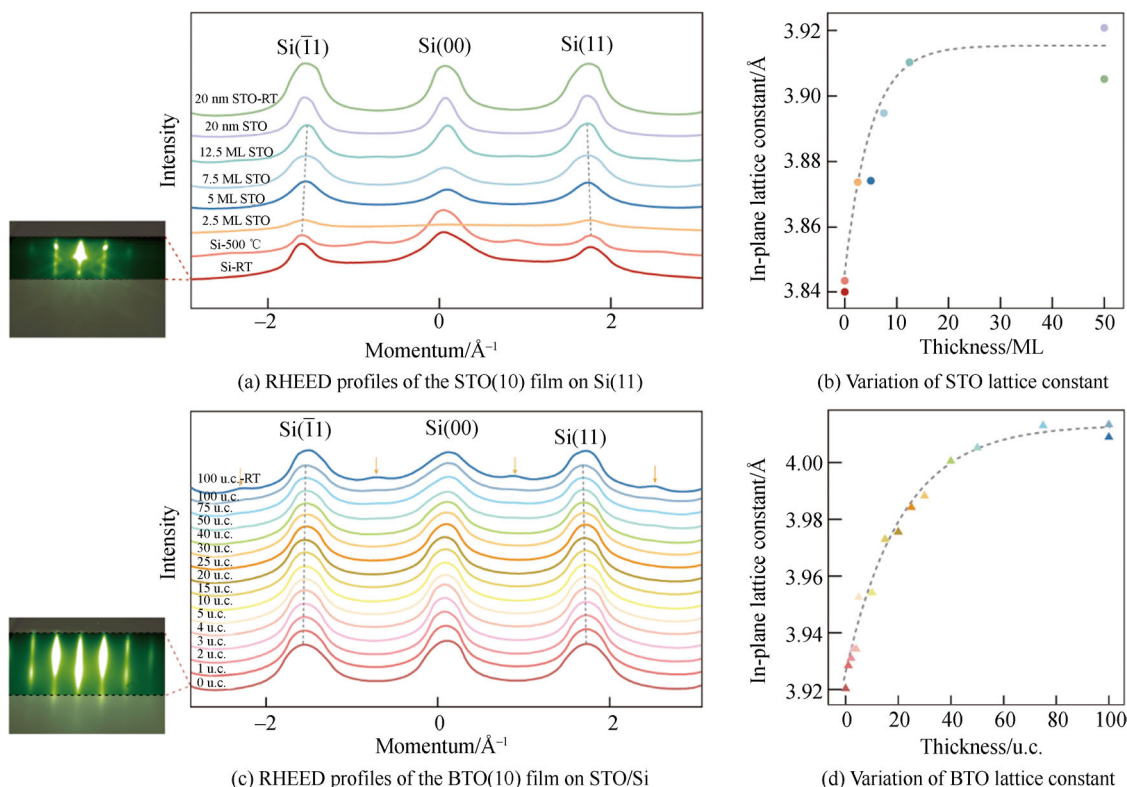


图3 STO 与 BTO 薄膜的 RHEED 衍射强度及面内晶格常数随薄膜厚度的演化趋势

Fig. 3 Variation of RHEED diffraction intensity and in-plane lattice constants with thickness during STO and BTO thin film growth

通过衍射峰位随时间的演化,可以观察到薄膜结晶质量与晶格弛豫的变化过程。在完成第 1 个 2.5 ML STO 的低温沉积高温退火流程后,STO(10)和 $(\bar{1}0)$ 衍射峰位相对 Si 衬底的(11)和 $(\bar{1}1)$ 峰位发生一定的偏移,此时 STO 的面内晶格常数为 3.874 Å,相比 Si 提供的有效晶格间距($5.431/\sqrt{2} = 3.840$ Å),薄膜已开始发生晶格弛豫,但衍射峰强度较弱,表明结晶质量有待改善。随着 STO 厚度增加,从 5 ML 开始到 12.5 ML(4.9 nm),衍射峰位进一步发生明显偏移,面内晶格常数从 3.874 Å 增大至 3.910 Å,标志着晶格弛豫持续发生,同时,峰强显著增强且峰形愈发尖锐,表明晶格弛豫发生过程伴随着结晶质量的显著提升。当 STO 厚度超过 12.5 ML 后,峰位和晶格常数没有进一步的明显变化,显示晶格已完成弛豫过程进入稳定状态。最终 20 nm 厚的 STO 薄膜在室温下的晶格常数为 3.905 Å,已接近块体 STO 的晶格参数。通过引入 20 nm STO 缓冲层,BTO 与衬底的晶格失配从直接外延时的 4.0% 有效降低至 2.2%,显著改善了 BTO 外延生长条件。

随后,通过同样的 RHEED 衍射强度曲线分析 BTO 的晶格弛豫过程。由峰位分析可知,在 STO/Si 基板上外延 BTO 薄膜,BTO 薄膜从第 1 个单胞层

开始观察到晶格弛豫现象,BTO 薄膜的面内晶格常数从 STO 提供的 3.905 Å 起始值快速弛豫至 3.928 Å,并随着厚度的增加持续弛豫,在 30 nm 厚度时达到 4.013 Å,并趋于稳定。随后在 40 nm BTO 薄膜从 700 °C 生长温度冷却至室温过程中,面内晶格常数从 4.013 Å 收缩至 4.009 Å。由于 BTO 薄膜受到来自 Si 衬底与 STO 的晶格约束,在升降温时不再严格遵循块体 BTO 薄膜的热膨胀规律。根据线性热膨胀关系 $a = a_0 \times [1 + \alpha \Delta T]$ ^[28],其中: a_0 为初始晶格常数; α 为线性热膨胀系数,计算得到 BTO 薄膜的有效热膨胀系数约为 $1.5 \times 10^{-6} \text{ K}^{-1}$,显著小于块体 BTO 薄膜的热膨胀系数(约为 $14.6 \times 10^{-6} \text{ K}^{-1}$),却与 Si 衬底的热膨胀系数($2.6 \times 10^{-6} \text{ K}^{-1}$)相近,表明 BTO 薄膜在冷却过程中受到 Si 衬底的热应变约束,从而在降温后保持较大的面内晶格常数,这对后续的极化调控起到了关键作用。此外,图 3c 的 RHEED 衍射谱中,箭头指向处出现了半阶峰,应为薄膜生长过程中常见的表面重构现象。

2.3 钛酸钡薄膜的晶体结构分析

图 4 为 300 nm BTO 薄膜的 X 射线衍射谱与界面的扫描透射电子显微镜照片。由图 4a 可见,XRD 谱 $2\theta-\omega$ 扫描结果显示,BTO 存在强度高且峰形尖

锐的衍射峰, 无杂相峰出现, 其中, 星号“*”对应 Si 衬底的衍射峰。通过峰位拟合计算得到面外晶格常数 $c = 3.988 \text{ \AA}$ 。BTO(002)衍射峰的摇摆曲线半峰宽为 0.45° (图 4a 左上角插图), 证实了 BTO 薄膜晶格的高度取向性。由图 4b 可见, BTO 薄膜的(202)衍射峰以及 Si 衬底的(101)衍射峰的 ϕ 扫描显示两者有 45° 偏差, 确认了 BTO 的[100]晶向沿 Si 的[110]晶向的明确外延关系, 与预期一致。倒易空间扫描映射(RSM)的定量分析(图 4c)显示, BTO 薄膜的面外晶格常数 $c = 3.990 \text{ \AA}$, 与 $2\theta-\omega$ 结果一致, 而面内晶格常数则为 $a = 4.010 \text{ \AA}$, 与前述 RHEED 数据的量化分析结果相印证。相对于块体 BTO, 薄膜的面内晶格常数 a 膨胀了 0.45% , 而 c 相对于块体收缩 1.2% , 从而得到薄膜的 $c/a = 0.994 < 1$, 与传统

块体 BTO 的 $c/a > 1.01$ 形成鲜明对比, 表明 BTO 薄膜中四方相的长轴倒向了面内。由于两者热膨胀系数的显著差异, 在冷却过程中 BTO 受到 STO 界面的约束, 无法自由收缩, 导致面内残留拉伸应力, 这种应力状态在面外方向产生压缩, 最终形成 $c/a < 1$ 的各向异性晶格畸变, 驱动极化从面外重取向至面内方向, 并延伸至整个 300 nm 厚的薄膜, 确保了宏观上的面内极化取向。

扫描透射电子显微镜(STEM)的分析结果进一步证实了 BTO 薄膜的高质量外延生长和清晰的 BTO/STO 界面结构(见图 4d)。除界面处几个 ML 的 STO 之外, 后续的 STO 和 BTO 薄膜都在高温高氧压下生长, 因而在 STO 与 Si 衬底的界面处形成了大约 4.5 nm 的 SiO₂ 非晶层。

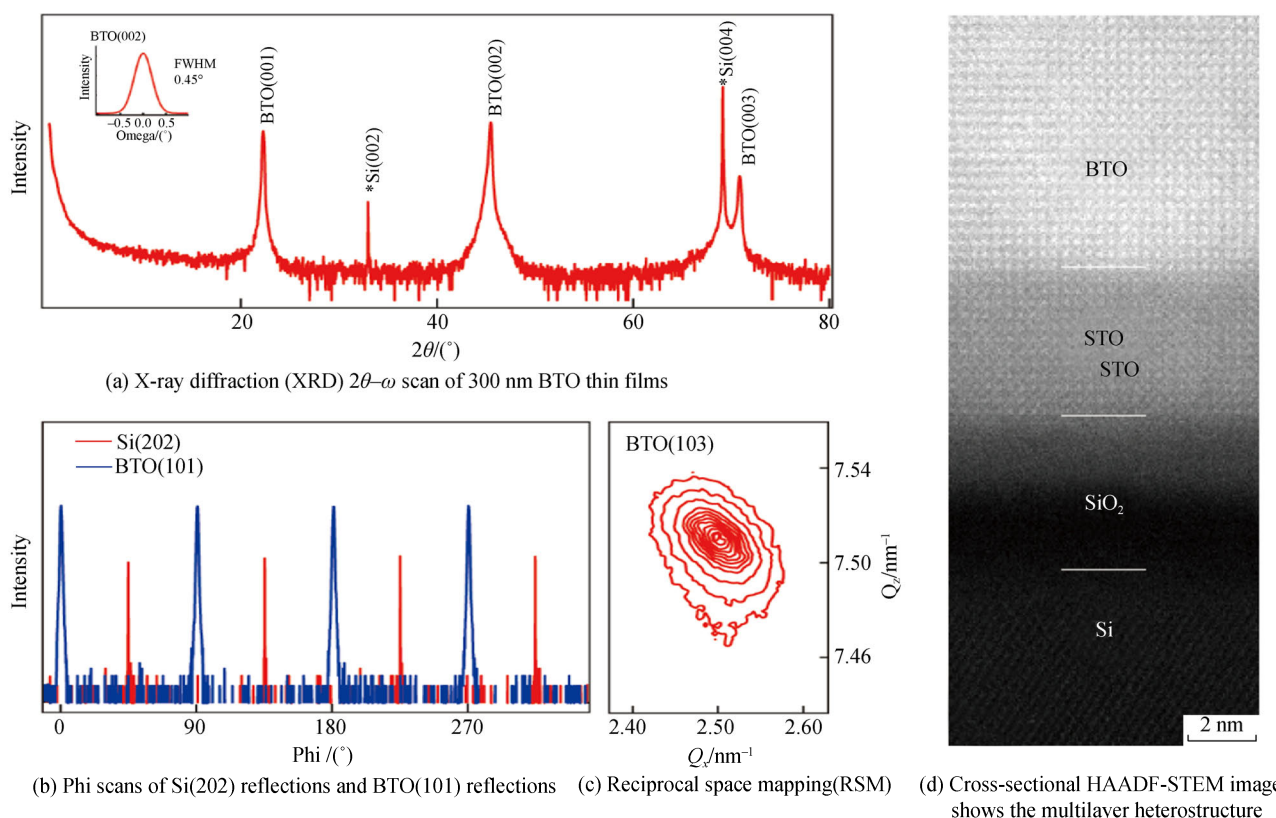


图 4 300 nm BTO 薄膜的 X 射线衍射谱与界面的扫描透射电子显微镜照片
Fig. 4 XRD characterization and STEM interfacial image of 300 nm BTO thin film

2.4 钛酸钡薄膜的铁电性质分析

压电力显微镜(PFM)能够在纳米尺度下直接观察铁电畴的分布和极化取向。采用横向 PFM(LPFM)和垂直 PFM(VPFM)模式对 BTO 薄膜的铁电特性进行了系统表征, 以验证 XRD 结构分析所揭示的面内极化取向。LPFM 和 VPFM 振幅图像展示了 BTO 薄膜中铁电畴的空间分布特征。

图 5 为 BTO 薄膜铁电畴的 PFM 表征结果。由图 5 可见, 2 种模式下均观察到明显的衬度变化, 表明样品具有明确的压电响应分布。LPFM 振幅信号强度显著高于 VPFM, 表明薄膜中存在显著的面内极化分量, 而面外极化分量较小, 且存在多种极化取向的混合状态, 进一步证实了结构中面内极化占主导地位。

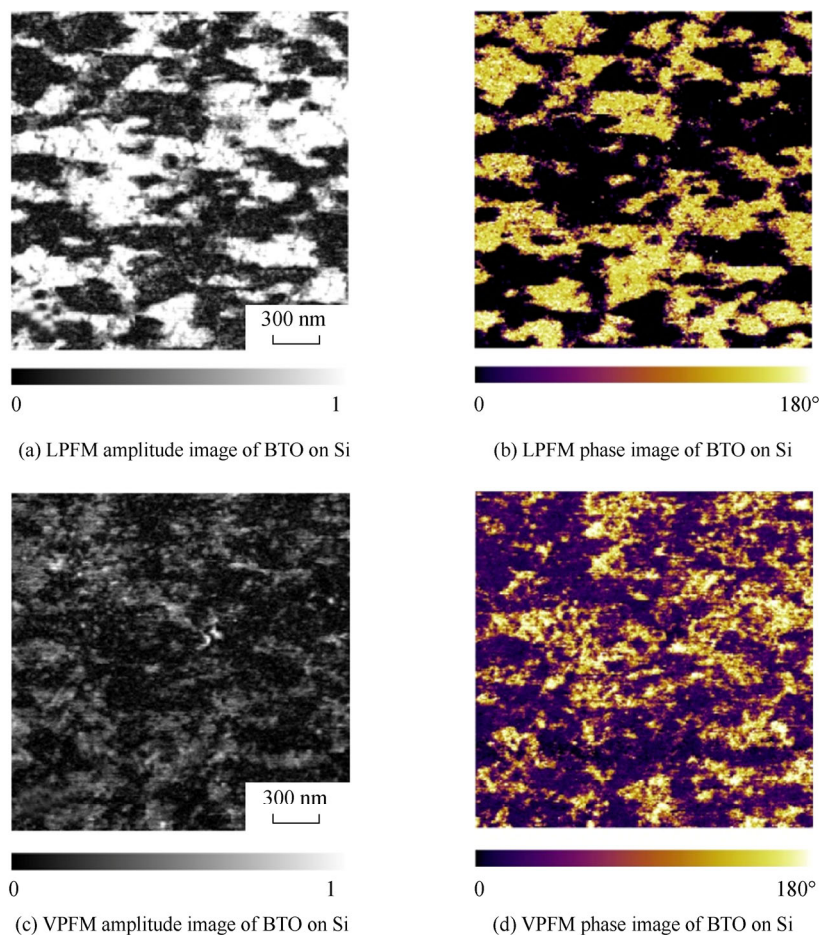


图 5 BTO 薄膜铁电畴的压电力显微镜(PFM)表征结果

Fig. 5 Piezoelectric force microscopy (PFM) characterization of ferroelectric domains in BTO thin films

3 结论

本工作通过 BME 技术成功实现了 2 in 晶圆级高质量 BTO 薄膜的硅基外延生长。通过原位 RHEED 监测技术,实时观测了生长过程中 STO 缓冲层和 BTO 薄膜的晶格弛豫过程,发现 STO 薄膜的晶格弛豫发生在界面到 12.5 ML 之间,而 BTO 的晶格弛豫过程则发生在界面到 30 nm 之间。基于这种晶格弛豫现象,STO 中间层为 BTO 提供了一个较低晶格失配度的模版,而 BTO 晶格发生弛豫则是最终获得面内极化薄膜的关键因素。原位 RHEED 数据分析还显示,受限于具有低热膨胀系数的 Si 衬底所提供的约束,BTO 薄膜在冷却至室温时仍保持较大的面内晶格常数,从而获得了面内极化占主导的稳定畴结构,这种原位监控技术为精确分析 BTO 薄膜的晶格弛豫过程,以及畴结构的控制提供了重要的信息,对推动硅基 BTO 新型电光调制器、高速通信器件、光量子计算芯片的研发具有重要意义。

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Lattice Relaxation and Domain Structure Engineering in BaTiO₃ Thin Films Epitaxially Grown on Silicon

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Extended Abstract

Introduction Silicon photonics has emerged as a critical technology for high-performance communication and computing, with electro-optic modulators serving as essential components. Barium titanate (BaTiO₃, BTO) exhibits exceptional promise due to its outstanding electro-optic coefficient ($r_{42} \approx 923$ pm/V) and compatibility with silicon-based fabrication processes. However, achieving high-performance BTO devices requires overcoming key materials integration challenges. The primary obstacles include the formation of amorphous SiO₂ interfacial layer during high-temperature, oxygen-rich BTO growth, which prevents effective epitaxy, and the substantial lattice mismatch between BTO ($a = b = 3.992$ Å, $c = 4.036$ Å) and Si (5.431 Å), resulting in approximately 4.0% mismatch even with 45° rotational epitaxy. This mismatch induces high-density dislocations and strain relaxation, compromising film quality. Strontium titanate (SrTiO₃, STO) buffer layers (3.905 Å) can reduce lattice mismatch to 1.7%, providing stress relief and serving as an epitaxial template. The electro-optic effect in BTO strongly depends on ferroelectric domain structure and crystallographic orientation, with in-plane polarization achieving maximum coefficients. While domain structures can be controlled through strain engineering and growth conditions, current analyses rely on post-growth characterization, lacking real-time monitoring of domain evolution processes and critical transition mechanisms.

Methods Oxide molecular beam epitaxy (MBE) combined with in situ reflection high-energy electron diffraction (RHEED) for real-time monitoring was employed to achieve controlled growth of high-quality BTO films at 2 in wafer scale. The systematic

investigation focused on lattice relaxation behavior during the growth of STO buffer layers and BTO layers with varying thicknesses. Silicon substrates underwent hydrofluoric acid (HF, 10% mass fraction) treatment to remove surface SiO₂ amorphous layers, followed by rapid transfer to ultra-high vacuum chambers (background pressure $\leq 5 \times 10^{-8}$ Pa). STO epitaxial growth employed a two-stage process: first, 2.5 monolayers (ML) of Sr and Ti metals were deposited at a substrate temperature of 200 °C under ultra-high vacuum ($< 5 \times 10^{-7}$ Pa) with precisely controlled oxygen partial pressure ($< 5 \times 10^{-6}$ Pa). The second stage involved in situ annealing at 500 °C for 8 minutes under ultra-high vacuum conditions ($< 1 \times 10^{-6}$ Pa). After three such cycles, additional STO layers were epitaxially grown using co-deposition at a thermocouple temperature of 600 °C under an oxygen partial pressure of 6×10^{-5} Pa. BTO layers were continuously co-deposited at 700 °C thermocouple temperature under 6×10^{-5} Pa oxygen partial pressure, ultimately obtaining 310 nm BTO/5 nm STO/Si heterostructures. Comprehensive characterization employed high-resolution four-circle X-ray diffraction (XRD) for 2θ - ω scanning and rocking curve analysis, spectroscopic ellipsometry for determining optical constants and thickness, atomic force microscopy (AFM) for surface morphology assessment, and piezoresponse force microscopy (PFM) for ferroelectric domain characterization.

Results and discussion Systematic RHEED diffraction pattern analysis during film growth enable quantitative analysis of lattice relaxation processes in both STO and BTO epitaxial films. STO buffer layers completed relaxation after 12.5 ML, reducing the lattice mismatch between BTO and Si from 4.0% to 2.2%. BTO films underwent progressive relaxation from the interface to a depth of 30 nm, with in-plane lattice constants evolving from 3.905 Å (STO-templated) to 4.013 Å at 30 nm, then stabilizing. During cooling from 700 °C to room temperature, thermal constraints from Si substrate significantly modified BTO behavior. The effective thermal expansion coefficient was $\sim 1.5 \times 10^{-6} \text{ K}^{-1}$, much lower than that of bulk BTO ($\sim 14.6 \times 10^{-6} \text{ K}^{-1}$) but comparable to Si ($2.6 \times 10^{-6} \text{ K}^{-1}$), indicating strong substrate constraint effects. The resulting 300 nm BTO films exhibited excellent quality: a rocking curve full width at half maximum (FWHM) of 0.45°, surface roughness < 0.2 nm, and critical lattice parameters $a = 4.010 \text{ Å}$, $c = 3.990 \text{ Å}$ ($c/a = 0.994 < 1$). This represents a 0.45% in-plane expansion and 1.2% out-of-plane contraction compared to bulk BTO, indicating a tetragonal phase reorientation with the long-axis in-plane. This anisotropic distortion creates residual tensile stress in-plane and compression stress out-of-plane, driving polarization reorientation from out-of-plane to in-plane throughout the entire film. PFM characterization confirmed this structure: lateral PFM (LPFM) signals significantly exceeded vertical PFM (VPFM) signals, demonstrating dominant in-plane polarization components. Optical characterization at 1550 nm revealed ideal performance with a refractive index $n = 2.28$ and an extinction coefficient $k \sim 10^{-6}$, indicating extremely low optical loss suitable for high-performance electro-optic modulators.

Conclusions High-quality BTO epitaxial growth on 2-inch silicon wafers was achieved via molecular beam epitaxy. In situ RHEED monitoring enabled real-time observation of lattice relaxation processes during STO buffer layer and BTO film growth, revealing that STO lattice relaxation occurs within the first 12.5 ML from the interface, while BTO lattice relaxation occurs within the first 30 nm from the interface. Based on this relaxation behavior, the STO intermediate layer provides BTO with a low-mismatch template, while BTO lattice relaxation is critical for ultimately achieving in-plane polarized films. In situ RHEED data analysis demonstrated that constraints from low thermal expansion coefficient Si substrate enable BTO films to maintain larger in-plane lattice constants during cooling to room temperature, thereby stabilizing domain structures with predominantly in-plane polarization. This in situ monitoring technique provides crucial information for precise analysis of BTO film lattice relaxation processes and domain engineering, offering significant implications for developing silicon-based BTO electro-optic modulators, high-speed communication devices, and optical quantum computing chips.

Keywords barium titanate; molecular beam epitaxy; silicon hetero-integration; strain engineering; in-plane ferroelectric polarization