Crystal Structure Characterization of MBE-grown Ultrathin TiN Epitaxial Films by Azimuthal Second Harmonic Generation

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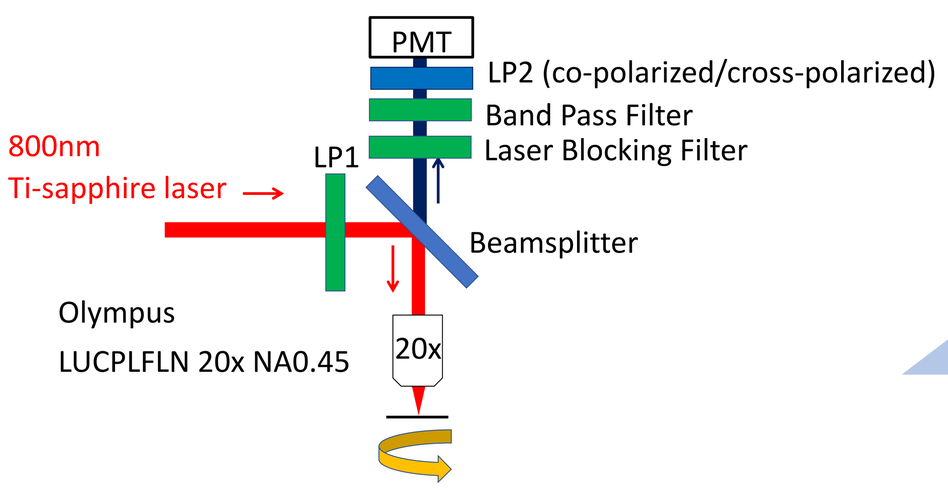
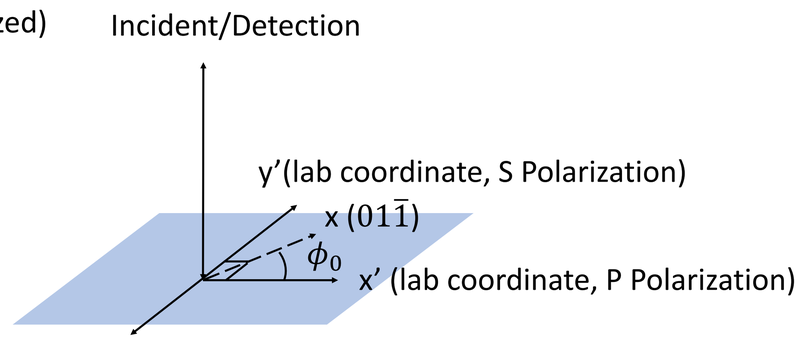
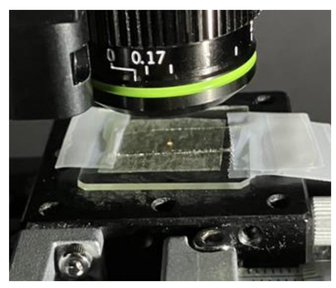
Abstract

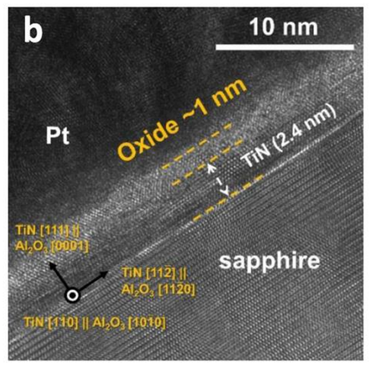
In this work we investigated the second harmonic generation (SHG) from the molecular-beam epitaxy (MBE) grown ultrathin TiN films on c-plane sapphire. We then performed the azimuthal second harmonic generation (AZSHG) measurement in normal incident configuration. The reflected AZSHG signal was measured in two distinguishable polarization configurations, where the polarization of the measured reflected SHG signal either parallel (i.e. co-polarized) or perpendicular (i.e. cross-polarized) with respective to the incident fundamental laser polarization. In this poster, we provide an interpretation about the observed AZSHG patterns and their intensity change by taking into account the SHG contributions from the epi-grown TiN surface, bulk, and its heterointerface with the sapphire substrate.

AZSHG Experimental Setup and Measurement









SEM showing 2.4nm TiN grown on c-plane Sapphire substrate [2]

- Nine TiN thin film samples with varied film thickness from 2nm to 46nm, were measured in this study.

Theory

For cubic centrosymmetric materials such as TiN, the SHG polarization due to surface and bulk contribution can be expressed as

$$P_{i\,(2\omega)} = \chi^{(2)}_{ijk} E_j\, E_k + \Gamma_{ijkl}\, E_j\, \nabla_k\, E_l$$

The first term is the electric-dipole term and can only come from the surface while the second term corresponding to electric-quadrupole/magnetic-dipole effects, give rise to the bulk contributed SHG. Both surface and bulk have anisotropic contributions. The combined SHG E-field from TiN(111) thin film has the phenomenological form of[1]

$$\mathrm{E}(2\omega) = \alpha - \beta \cos 3\phi$$

Isotropic term, contributed by the surface, bulk and interface between TiN and sapphire substrate.

Anisotropic term, depends on $\chi^{(2)}_{xxy}$ of the TiN(111) surface structure (*3m* crystal class) and bulk contribution.

Furthermore, we find that an additional isotropic term γ was necessary in the fitting formula to quantify the isotropic AZSHG curves observed in the plotted AZSHG curves shown in the right column.

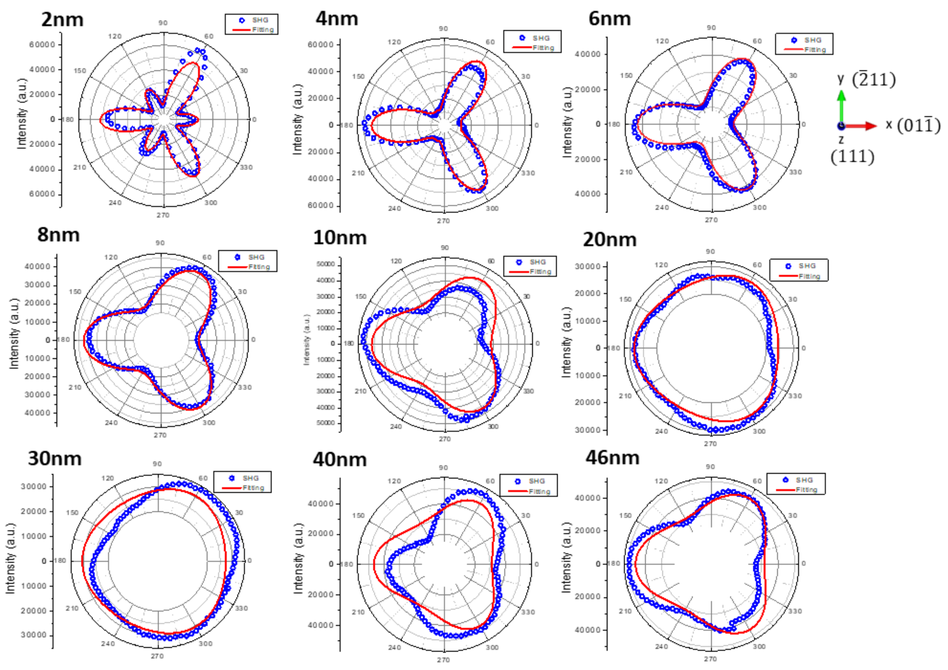
Co-polarized: $\mathrm{I}(2\omega) = |\alpha - \beta \cos(3(\phi - \phi_0))|^2 + \gamma^2$

Cross-polarized: $\mathrm{I}(2\omega) = |\alpha + \beta \sin(3(\phi - \phi_0))|^2 + \gamma^2$

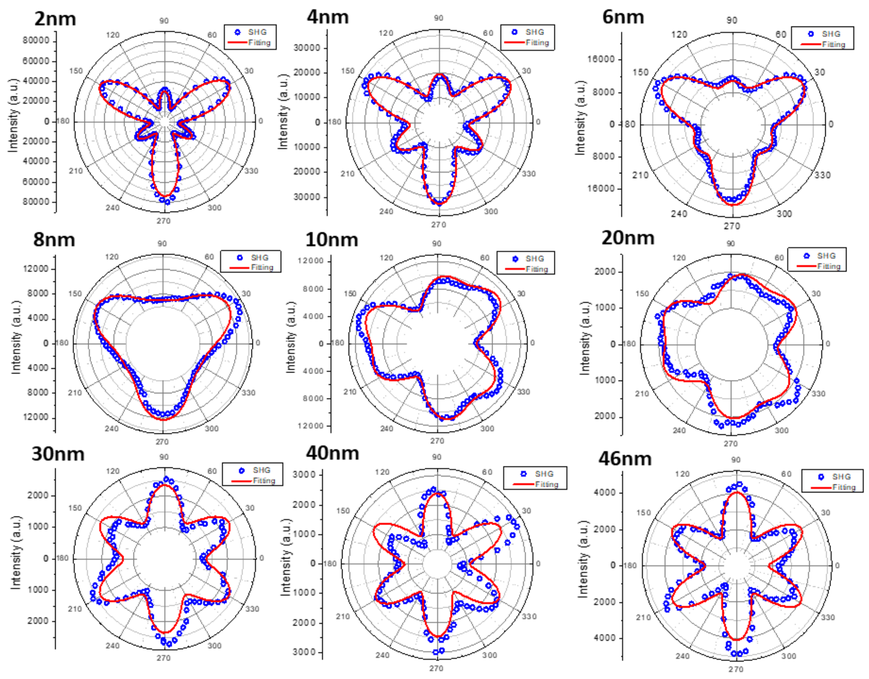
ϕ_0 : defines the initial angle between the incident E field and crystal x $(01\bar{1})$ axis

Results

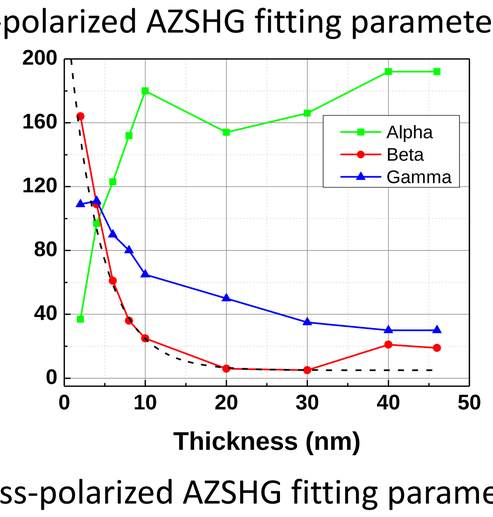
Co-polarized AZSHG:



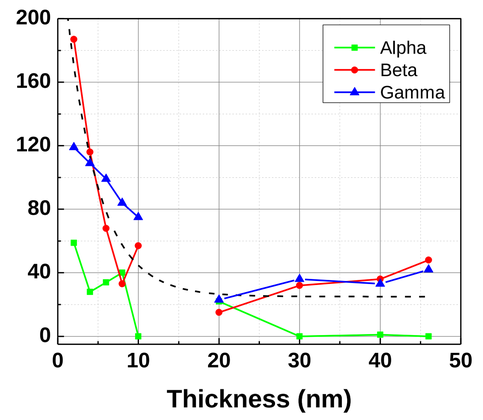
Cross-polarized AZSHG:



Co-polarized AZSHG fitting parameters



Cross-polarized AZSHG fitting parameters



- Surface contributed AZSHG governs the six-fold symmetry pattern (β term).
- In ultrathin TiN samples, the combination of surface and interface contributed SHG give rise to the three-fold symmetry pattern in both polarization configuration.
- For thicker TiN samples, the heterointerface contributed SHG is diminished and the bulk and surface contributed SHG dominates the observed AZSHG pattern.

Discussion & Conclusion

- The additional isotropic term γ diminished as the TiN film thickness increases. We can attribute this term to the SHG contributed from the lattice mismatch between the TiN/sapphire heterointerface.
- In ultrathin TiN sample, the combination of surface and heterointerface SHG gives the feature of three-fold symmetry AZSHG in both polarization configuration.
- The bulk contributed AZSHG, due to electric quadrupole effect, dominates the total AZSHG in thick TiN samples (note: increasing α/β ratio). This effect is only observable in co-polarization configuration (note: increasing α term in co-polarization).
- Surface contributed AZSHG diminished when the film thickness is increasing (note: decreasing β in both polarization configuration).
- In both polarization configuration, the isotropic term γ do not diminished completely in thicker sample. The origin of this isotropic AZSHG signal need to be further investigated in the future.

Reference:
1. Appl. Phys. A 59, 545-552 (1994)
2. 2. ACS Appl. Mater. Interfaces 2022, 14, 16839-16845

Acknowledgement:
The work done at National Taiwan University is supported by the Ministry of Science and Technology under the following funding grants: MOST 111-2112-M-002 -035 -MY3, NSTC 111-2119-M-002-013 -MBK, and NSTC 112-2811-M-002-078.