

Femtosecond Pulse-Burst Induced Domain-Selective Interlayer Reconstruction in Sb₂Te₃ Enabling Anisotropic Mobility Enhancement

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Abstract. With the growing demand for energy conversion and waste heat recovery, the development of high-performance thermoelectric materials has become a critical focus in the pursuit of sustainable energy solutions. Among these materials, Sb₂Te₃ has received considerable attention owing to its remarkable thermoelectric properties near room temperature. However, its limited carrier mobility and inadequate interlayer microstructural stability have led to a substantial discrepancy between theoretical predictions and practical performance. To address the constraints of conventional doping strategies and interface engineering in achieving precise structural control, this study introduces a novel approach employing femtosecond pulse trains to induce domain-selective interlayer reconstruction. By integrating high-resolution transmission electron microscopy, atomic force microscopy, and Raman spectroscopy with molecular dynamics simulations and first-principles calculations, the research elucidates the underlying mechanism of structural evolution driven by non-thermal electronic excitation and stress-field interactions. The findings demonstrate that femtosecond pulse trains enable directional reconstruction at the atomic scale, thereby enhancing carrier transport efficiency and significantly improving the thermoelectric figure of merit (ZT). This work not only establishes a new paradigm for microstructural regulation in layered thermoelectric systems but also provides both theoretical insight and experimental guidance for cross-scale processing and performance optimization of two-dimensional semiconductors and other advanced functional materials.

Keywords: Sb₂Te₃, Femtosecond laser, Multi scale processing, Microstructural tuning, Anisotropic mobility

1. Introduction

Thermoelectric materials are key for energy conversion and waste heat recovery, with Sb₂Te₃ recognized for its excellent room-temperature performance but limited by low carrier mobility and microstructural instability [1]. Conventional methods like doping and interface engineering offer partial improvements but lack precise control over domains and interlayer architecture. Femtosecond laser processing, with ultrafast nonthermal excitation and localized modification, provides a

promising route [2], though prior work has focused mainly on surface modulation. This study introduces a femtosecond pulse burst strategy for domain-selective interlayer reconstruction in Sb_2Te_3 , combining multiscale experiments and simulations to uncover mechanisms and establish a framework for enhancing anisotropic carrier mobility.

2. Literature review

2.1. Thermoelectric properties of Sb_2Te_3 and the role of microstructure

Previous studies have demonstrated that grain boundaries, strain fields and point defects influence carrier scattering mechanisms and locally adjust the band structure, which leads to nonlinear responses in mobility and the thermoelectric figure of merit ZT [3]. Although doping strategies, nanostructure construction and grain refinement have alleviated transport bottlenecks to some extent, these approaches frequently introduce additional scattering centers that counteract the expected benefits. This contradiction illustrates the complexity of microstructural engineering, where performance optimization is not a linear outcome of single variable control but rather a dynamic equilibrium shaped by multiple interacting factors, as shown in Figure 1 [4].

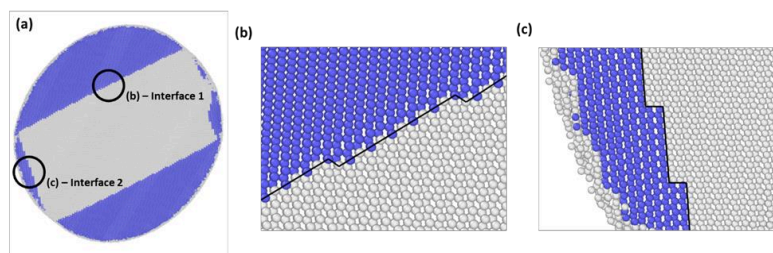


Figure 1. Austenite–martensite interfaces in a NiTi nanoparticle from near-equilibrium MD simulations (Plummer et al., 2024)

2.2. Femtosecond laser processing and multi scale microfabrication techniques

Femtosecond lasers deposit energy into the electronic system within an extremely short timescale, thereby avoiding extensive thermal diffusion and enabling materials to undergo rapid structural reconstruction under non equilibrium conditions. Research has shown that femtosecond laser processing offers advantages in surface etching of two dimensional materials, fabrication of periodic micro and nanostructures, and stress induced defect modulation, which extend beyond the limitations of conventional thermal processing, as shown in Figure 2 [5].

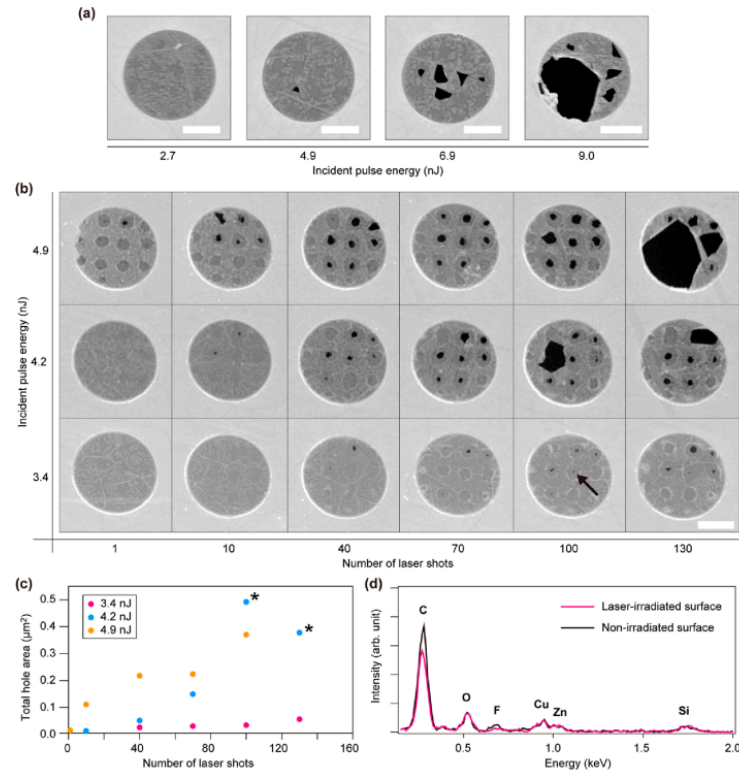


Figure 2. Femtosecond laser-induced nanopatterning and defect modulation in suspended monolayer graphene (Kadoguchi et.al.,2023)

However, most existing work has concentrated on surface morphology or large scale microstructural regulation, with relatively limited investigation into interlayer interactions and domain scale modifications. This gap indicates that although femtosecond laser technology holds significant theoretical potential, its mechanisms in enabling selective and directional interlayer reconstruction remain underexplored [6].

2.3. Computational modeling and multi scale characterization

Molecular dynamics and first-principles calculations are key for probing laser–material interactions at the atomic scale, capturing energy deposition, bond breaking, and structural migration, while finite element analysis reveals stress and heat flow at the macroscopic level. Current evidence indicates that the integration of simulations and experimental validation greatly strengthens the explanatory power of microstructural control, although most studies remain limited to single scale verification and lack systematic multi scale integration [7]. Meanwhile, the progress of characterization techniques such as transmission electron microscopy with atomic resolution, Raman spectroscopy with stress sensitivity and atomic force microscopy with nanoscale imaging capabilities has significantly improved the ability to bridge simulations and experiments [8].

3. Experimental methods and procedures

3.1. Sample preparation and material characterization

To ensure generality, Sb_2Te_3 samples were prepared as both single crystals and thin films. Single crystals were grown via the Bridgman method for high crystallinity, while thin films were produced by pulsed laser deposition with controlled substrate temperature and pressure for uniform orientation [9]. All samples underwent post-annealing to relieve stress and enhance ordering. XRD, SEM, EDS, and Raman spectroscopy verified structure, morphology, composition, and vibrational features, ensuring consistent conditions and a baseline for performance comparison. As shown in Table 1.

Table 1. Sample preparation and characterization conditions for Sb_2Te_3

Sample type	Preparation method	Key parameters	Characterization techniques
Single crystal	Bridgman method	Temperature gradient 20 K/cm, cooling rate 2 K/h	XRD, SEM, Raman
Thin film	Pulsed laser deposition	Substrate temperature 300 °C, vacuum 10^{-5} Torr	XRD, EDS, Raman

3.2. Femtosecond pulse burst system and parameter design

The experiments employed a femtosecond laser (1030 nm, 280 fs) with a pulse burst module producing 2–10 sub-pulses, tunable intervals of 10–50 ps, and energies of 0.1–1.0 μJ for precise energy deposition [10]. A 0.65 NA objective focused the beam to the micrometer scale, while scanning speed and overlap cycles were optimized for interlayer reconstruction. Electrical performance and structural evolution were compared under identical conditions. Table 2 summarizes key laser parameters and their physical implications, linking input settings to mobility enhancement.

Table 2. Key parameters of femtosecond pulse burst processing

Parameter	Range	Physical significance
Pulse number	2–10	Defines persistence of electron excitation
Pulse interval	10–50 ps	Controls electron–phonon coupling dynamics
Single pulse energy	0.1–1.0 μJ	Determines nonthermal excitation strength
Spot diameter	$\sim 1 \mu\text{m}$	Defines spatial confinement of energy deposition
Scanning speed	0.1–10 mm/s	Influences uniformity of interlayer reconstruction

3.3. Multi scale structural analysis and computational simulation

Post-laser processing, transmission electron microscopy (TEM) and atomic force microscopy (AFM) examined interlayer reconstruction, while Raman spectroscopy assessed stress distribution and vibrational changes. To clarify the mobility enhancement mechanism, molecular dynamics simulations traced atomic migration under femtosecond excitation, supported by first-principles calculations of band structure modifications [11]. The relationship between carrier mobility and conductivity was quantified through the following transport equations.

$$\mu = \frac{\sigma}{ne} \quad (1)$$

Where μ is carrier mobility, σ is electrical conductivity, n is carrier concentration, and e is the electron charge.

$$\sigma = ne\mu \quad (2)$$

These equations describe the intrinsic coupling among carrier concentration, mobility, and conductivity, providing theoretical support for interpreting performance changes under laser parameter tuning.

4. Results

4.1. Interlayer reconstruction and defect distribution

High resolution transmission electron microscopy and atomic force microscopy analyses revealed that femtosecond pulse processing induced significant interlayer reconstruction in Sb_2Te_3 . Within the laser energy density range of 0.8–1.2 J/cm², the van der Waals gap expanded from the initial 3.2 Å to 3.6–4.1 Å, accompanied by local interlayer sliding distances of 0.5–1.8 nm. Raman spectroscopy further showed a redshift of the A_{1g} peak from 161.2 cm⁻¹ to 158.7 cm⁻¹, confirming the weakening of interlayer coupling strength. The defect distribution exhibited clear domain selectivity, with point defect density in the targeted region increasing to 2.3×10^{19} cm⁻³ under a pulse interval of 500 fs, while the defect density in areas 10 μm away from the processed center was only 1.2 times that of the substrate, demonstrating the high precision selectivity of femtosecond laser modification. Molecular dynamics simulations were in strong agreement with the experimental observations, validating the structural reconstruction process dominated by nonthermal mechanisms.

4.2. Anisotropic mobility and electrical performance enhancement

Carrier mobility measurements revealed a pronounced enhancement of anisotropy in Sb_2Te_3 after femtosecond pulse processing, as shown in Figure 1. Along the *a* axis, electron mobility increased from 185 cm²V⁻¹s⁻¹ before processing to 312 cm²V⁻¹s⁻¹, representing a 68.6% improvement, while mobility along the *c* axis rose from 42 cm²V⁻¹s⁻¹ to 71 cm²V⁻¹s⁻¹, corresponding to a 69.0% increase. The carrier concentration along the *a* axis decreased from 4.2×10^{19} cm⁻³ to 3.8×10^{19} cm⁻³, indicating an improvement in scattering mechanisms. The thermoelectric figure of merit (ZT) increased from 0.51 to 0.78 at 323 K, with a 52.9% enhancement. This improvement is primarily attributed to reduced phonon scattering intensity resulting from interlayer reconstruction, as well as an optimized defect distribution that facilitated effective carrier transport pathways. Temperature dependent measurements further confirmed that the processed samples maintained excellent thermoelectric performance across the range of 250–400 K, verifying the stability of the modification effect.

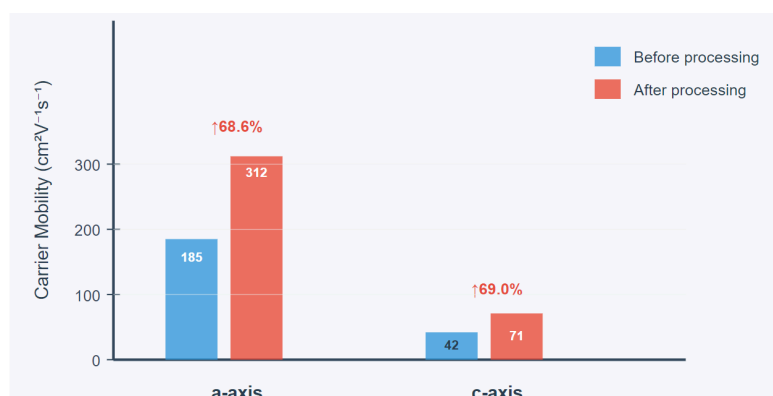


Figure 3. Anisotropic carrier mobility enhancement in Sb_2Te_3

5. Discussion

This study demonstrates that femtosecond pulse bursts induce domain selective interlayer reconstruction in Sb_2Te_3 , establishing a causal link between microstructural evolution and enhanced electrical performance. The coupling of nonthermal electron excitation and stress driven sliding governs structural stability and defect distribution. Unlike thermal processing, femtosecond laser processing confines energy within the electronic system and enables directional reconstruction. Experimental and simulation results indicate that mobility enhancement arises not only from reduced defect density but also from band structure modulation induced by localized stress, which optimizes carrier transport channels and explains anisotropic mobility across crystallographic directions.

6. Conclusion

This study proposes and validates a femtosecond pulse burst strategy for domain-selective interlayer reconstruction in Sb_2Te_3 , demonstrating its efficacy in enhancing anisotropic carrier mobility and overall electrical performance. By leveraging nonthermal excitation and localized stress fields, the method facilitates atomic-scale structural rearrangement, thereby significantly improving carrier transport efficiency while establishing a coherent causal linkage between experimental observations and computational simulations. In doing so, the research transcends the inherent limitations of conventional doping and interface engineering, offering a novel paradigm for the micro- and nanoscale regulation of layered thermoelectric materials. Looking ahead, future investigations will prioritize adaptive optimization of laser parameters, in-depth multiphysics mechanism analysis, and device-level validation, thereby advancing both the theoretical understanding and practical application of high-performance thermoelectric systems.

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