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$(Bi_{0\cdot5}Sb_{0\cdot5})_2Te_3$ PLYONKALARINING CHASTOTAGA BOG‘LIQ TENZOREZISTIV XOSSALARI: TSIKLIK DEFORMATSIYA TA’SIRI

Annotatsiya. Kompozit $(Bi_{0\cdot5}Sb_{0\cdot5})_2Te_3$ yupqa plyonkalarining chastotaga bog'liq tenzorezistiv xossalari siklik mexanik deformatsiya sharoitida tadqiq qilindi. Plyonkalar vakuumli termik bug'lantirish usuli bilan tayyorlanib, navbatma-navbat deformatsiya tsikllariga uchratildi. Kritik chastota sezilarli darajada kamayishi kuzatildi, bu esa kristallitlar orasida dielektrik bo'shliqlarning hosil bo'lishi va inter-kristallit kontaktlarning yemirilishi bilan izohlanadi. Elektr qarshiligining chastotaga bog'liqligi ekvivalent elektr sxemasi modeli yordamida tushuntirildi, bu modelda hajmiy qarshilik, kontakt qarshiligi va shuntlovchi sig'im mavjud. Tenzorezistiv sezgirlikning yuqoriligi va deformatsiya xarakteristikalarining nochiziqligi ushbu plyonkalarni mexanik charchashni aniqlovchi datchiklar va moslashuvchan elektron qurilmalar uchun istiqbolli ekanligini ko'rsatadi.

Kalit so‘zlar: tenzorezistiv xossalar; $(Bi,Sb)_2Te_3$ yupqa plonkalar; siklik deformatsiya; kontakt qarshiligi; mexanik charchashni aniqlash

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ЧАСТОТНАЯ ЗАВИСИМОСТЬ ТЕНЗОРЕЗИСТИВНОГО ПОВЕДЕНИЯ ПЛЕНОК $(Bi_{0\cdot5}Sb_{0\cdot5})_2Te_3$: ЭФФЕКТЫ ЦИКЛИЧЕСКОЙ ДЕФОРМАЦИИ

Аннотация. Исследованы частотные зависимости тензорезистивных свойств композитных тонких плёнок $(Bi_{0\cdot5}Sb_{0\cdot5})_2Te_3$ при циклической механической деформации. Плёнки были получены методом вакуумного термического испарения и подвергнуты циклическим деформациям. Было наблюдено значительное снижение критической частоты, что связано с образованием диэлектрических зазоров и деградацией межкристаллитных контактов. Частотная зависимость электрического сопротивления была объяснена с использованием эквивалентной электрической схемы, включая объёмное сопротивление, контактное сопротивление и шунтирующую ёмкость. Высокая тензорезистивная чувствительность и нелинейные характеристики деформации показывают потенциал этих плёнок для использования в датчиках накопленного усталостного повреждения и гибкой электроники.

Ключевые слова: тензорезистивные свойства; тонкие плёнки $(Bi,Sb)_2Te_3$; циклическая деформация; контактное сопротивление; диагностика усталостных повреждений;

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FREQUENCY-DEPENDENT TENSORESISTIVE BEHAVIOR OF $(Bi_{0\cdot5}Sb_{0\cdot5})_2Te_3$ FILMS: CYCLIC DEFORMATION EFFECTS

Abstract. The frequency-dependent tensoresistive properties of composite $(Bi_{0\cdot5}Sb_{0\cdot5})_2Te_3$ thin films were investigated under cyclic mechanical deformation. The films were fabricated by vacuum thermal evaporation and subjected to alternating deformation cycles. A significant decrease in critical frequency was observed, linked to the formation of dielectric gaps and the degradation of inter-grain contacts. The frequency dependence of electrical resistance was explained using an equivalent circuit model, incorporating bulk resistance, contact resistance, and shunting capacitance. The high tensoresistive sensitivity and nonlinear deformation characteristics indicate the potential for these films to be used in fatigue-damage sensing and flexible electronic applications.

Keywords: tensoresistive properties; $(Bi,Sb)_2Te_3$ thin films; frequency dependence; contact resistance; fatigue damage sensing;

Introduction

Bismuth-antimony telluride compounds, particularly $(Bi_{0,5}Sb_{0,5})_2Te_3$, represent a critical class of thermoelectric materials with exceptional performance near room temperature. Recent advances have demonstrated their potential not only for energy conversion but also for multifunctional sensing applications [1]. The coupling between mechanical deformation and electrical properties-known as the tensoresistive or piezoresistive effect-has attracted considerable attention for developing flexible thermoelectric devices and strain sensors [2]. Polycrystalline semiconductor films exhibit inherent non-homogeneity arising from grain boundaries, crystallite interfaces, and microstructural defects. These features profoundly influence electrical transport properties, particularly under mechanical stress. Understanding the frequency-dependent electrical behavior of such films is essential for optimizing their performance in dynamic operating conditions and for elucidating the fundamental mechanisms governing charge transport across grain boundaries [3].

Percolation theory provides a powerful framework for interpreting the electrical behavior of composite and polycrystalline materials near critical thresholds [4]. In the context of thin films, percolation models can explain the transition from insulating to conducting behavior and the emergence of frequency-dependent conductivity as conductive pathways form or break under mechanical deformation [5]. Despite extensive research on thermoelectric properties of Bi_2Te_3-based materials [6], the combined effects of cyclic mechanical deformation and AC frequency on tensoresistive behavior remain incompletely understood. This study addresses this gap by systematically investigating how cyclic deformation alters the frequency-dependent electrical characteristics of $(Bi_{0,5}Sb_{0,5})_2Te_3$ composite films and by employing percolation theory to explain the observed high tensoresistance coefficients and nonlinear deformation characteristics.

Materials and methods

Composite $(Bi_{0,5}Sb_{0,5})_2Te_3$ films were fabricated using vacuum thermal evaporation, a technique widely employed for producing high-quality thermoelectric thin films [7]. Two sets of films were prepared at different substrate temperatures: T_{sub}=150°C and T_{sub}=90°C. The substrate temperature critically influences film microstructure, porosity, and grain boundary characteristics, thereby affecting both thermoelectric and mechanical properties [8].

Selected film samples were subjected to cyclic alternating deformations with an amplitude of $\varepsilon = \pm 1\times10^{-3}$ (relative strain units) for $N = 5\times10^{5}$ cycles. This cyclic loading protocol simulates realistic operational conditions for flexible thermoelectric devices and strain sensors, which must withstand repeated mechanical stress [9]. The deformation was applied in a controlled manner to ensure reproducibility and to avoid catastrophic failure of the films. Electrical resistance measurements were performed as a function of AC frequency over a broad range from DC (0 Hz) to 10 Hz, with extended measurements up to 100 kHz-1 MHz for selected samples. This wide frequency range enables the identification of critical frequencies and the characterization of frequency-dependent transport mechanisms [10]. Measurements were conducted at room temperature using a precision impedance analyzer.

An equivalent circuit model was developed to interpret the frequency-dependent electrical behavior. The model consists of: - R_v: Crystallite resistance (bulk resistance of individual grains) - R_c: Contact resistance between adjacent crystallites – C_c: Capacitance associated with dielectric gaps at grain boundaries, shunting R_c

This circuit topology reflects the microblock structure of polycrystalline films and has been successfully applied to similar composite systems [11]. The critical frequency ω_c is determined by the R_C time constant: $\omega_c \approx \frac{1}{R_c C_c}$

Percolation theory was employed to explain the high tensoresistance coefficient and the nonlinearity of static deformation characteristics. In percolation models, the electrical conductivity near the percolation threshold follows a power-law relationship with the volume fraction of conductive phase. For polycrystalline films under mechanical stress, changes in contact resistance and the formation or disruption of conductive pathways can be interpreted within this framework [12].

Results

Figure 1 presents the frequency dependence of electrical resistance for *$(Bi_{0,5}Sb_{0,5})_2Te_3$* films under different preparation and treatment conditions. Three distinct behaviors are observed:

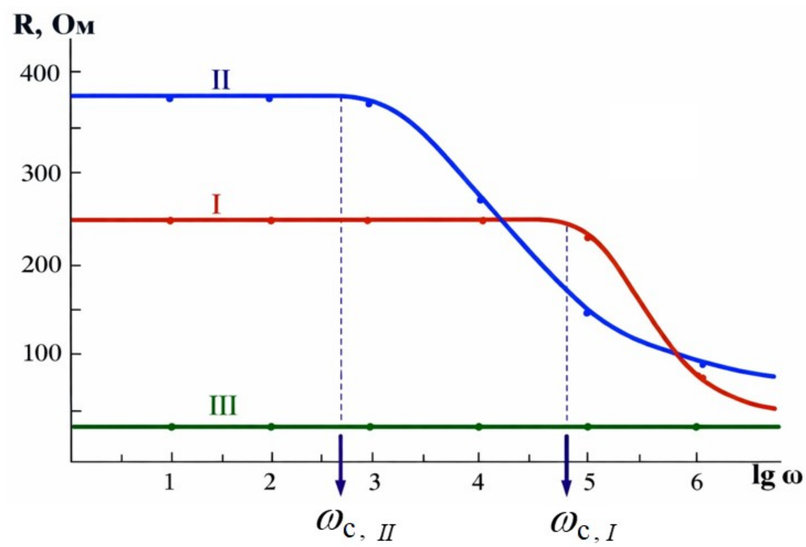

Figure 1. Dependence of the film resistance on the frequency of the supply voltage: curve I-before exposure, curve II-after exposure to $N = 5 \times 10^5$ deformation cycles for films with $T_h = 90^\circ C$; curve III-$T_h = 150^\circ C$.

Curve I (films prepared at T_{sub} = 90°C before cyclic deformation): These films exhibit a sharp decrease in resistance with increasing frequency, starting from a threshold frequency. The relatively high porosity and weak inter-grain contacts in films prepared at lower substrate temperatures result in significant contact resistance R_c and associated capacitance C_c, leading to pronounced frequency dependence [13].

Curve II (films prepared at T_{sub} = 90°C after N = 5×10^5 cycles): After cyclic deformation, the critical frequency ω_c decreased by approximately two orders of magnitude ($\omega_{c1}/\omega_{cd} \approx 10^2$). This dramatic shift indicates a substantial increase in the shunting capacitance C_c, attributed to the formation of new dielectric gaps between crystallites due to accumulated mechanical fatigue [14].

Curve III (films prepared at T_{sub} = 150°C): Films prepared at higher substrate temperatures show nearly frequency-independent resistance across the measured range. The denser microstructure and stronger inter-grain bonding result in lower contact resistance and reduced capacitive effects, consistent with observations in other high-quality thermoelectric films.

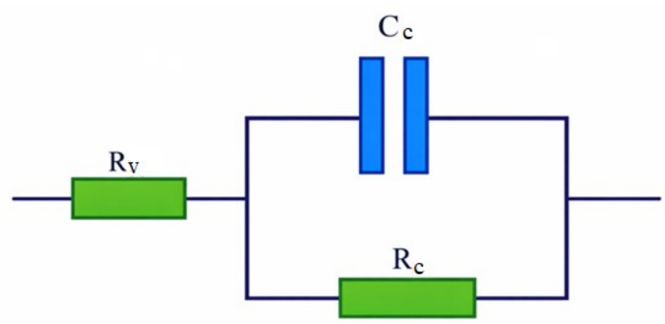

Figure 2. Equivalent resistance circuit of a microblock of a ¿ film.

Figure 2 illustrates the equivalent circuit model for a microblock within the polycrystalline film. The model successfully captures the observed frequency dependence: at low frequencies ($\omega << \omega_c$), the capacitance C_k acts as an open circuit, and the total resistance is R_v+R_c. At high frequencies ($\omega>>\omega_c$), C_c effectively short-circuits R_c, reducing the total resistance to approximately R_v.

The quantitative agreement between the model predictions and experimental data (Figure 1) validates the proposed microstructural interpretation. The shift in critical frequency after cyclic deformation can be quantitatively explained by the increase in C_c due to the widening of dielectric gaps at grain boundaries [15].

The tensoresistive response of the films depends strongly on the type of deformation: Tensile deformation (stretching): Increases the separation between crystallites, thereby increasing contact resistance R_c and the total film resistance. This behavior is consistent with the negative piezoresistance observed in Bi_2Te_3 nanoparticle-based fabrics under lateral strain [16].

Compressive deformation: Decreases the inter-grain spacing, enhancing electrical contacts and reducing film resistance. This asymmetric response contributes to the nonlinearity of the static deformation characteristics. The high tensoresistance coefficient observed in these films can be attributed to the sensitivity of contact resistance to mechanical stress, a phenomenon well-explained by percolation theory near the percolation threshold.

Discussion

The application of percolation theory to polycrystalline $(Bi_{0,5}Sb_{0,5})_2Te_3$ films provides a coherent explanation for the observed high tensoresistance coefficients and nonlinear deformation characteristics. In percolation models, the electrical conductivity σ near the percolation threshold follows: $\sigma \propto (\varphi - \varphi_c)^{\lambda}$ where φ is the volume fraction of the conductive phase, φ_c is the percolation threshold, and t is the critical exponent (typically t≈2–3 for three-dimensional systems). In polycrystalline films, mechanical deformation effectively modulates φ by altering the connectivity of conductive pathways through changes in contact resistance [17].

Recent studies on carbon nanotube/epoxy composites and graphene-based strain sensors have demonstrated similar percolation-driven tensoresistive behavior, with gauge factors and sensitivity strongly dependent on proximity to the percolation threshold. The transition from linear to nonlinear piezoresistive behavior observed in thermally reduced graphene oxide under large strains (>7.5%) further supports the percolation interpretation [18]. The dramatic decrease in critical frequency after 5×10^5 deformation cycles reflects the accumulation of mechanical fatigue damage in the film microstructure. Cyclic loading induces progressive degradation of inter-grain contacts, formation of micro-cracks, and widening of dielectric gaps. These structural changes increase the capacitance C_c while potentially increasing contact resistance R_c, shifting the critical frequency to lower values.

This sensitivity to cyclic deformation suggests a promising application for $(Bi,Sb)_2Te_3$ films as sensors for accumulating fatigue damage in structural health monitoring. Similar concepts have been explored in carbon nanotube/epoxy nanocomposites, where electrical impedance analysis under cyclic loading provides insights into material degradation. The ability to detect fatigue damage through changes in frequency-dependent electrical properties could enable real-time monitoring of critical aerospace and civil engineering structures [19].

The frequency-dependent electrical behavior and tensoresistive properties of $(Bi_{0,5}Sb_{0,5})_2Te_3$ films have important implications for flexible thermoelectric devices. Recent advances in flexible Bi_2Te_3-based films have achieved impressive thermoelectric performance with power factors exceeding 1000 $\mu W\ m^{-1}\ K^{-2}$ and excellent mechanical durability (less than 10% degradation after 700–1000 bending cycles) [20], [21]. However, the present study reveals that even when macroscopic electrical properties remain stable, microstructural changes induced by cyclic deformation can significantly alter frequency-dependent characteristics. This finding is particularly relevant for applications involving AC power generation or high-frequency operation, where the effective resistance may differ substantially from DC measurements.

The multifunctional capabilities of Bi_2Te_3-based materials—combining thermoelectric energy conversion with strain, pressure, and temperature sensing—have been demonstrated in recent stretchable fabrics and flexible films [22]. The integration of percolation-based tensoresistive sensing with thermoelectric functionality could enable self-powered structural health monitoring systems, where the same material simultaneously harvests energy and detects mechanical damage [23].

The observed frequency-dependent conductivity in $(Bi_{0,5}Sb_{0,5})_2Te_3$ films aligns with broader trends in composite materials near the percolation threshold. Studies on epoxy/graphite flake microcomposites have shown that at the percolation threshold (0.5 wt.% graphite flakes), significant increases in permittivity, conductivity, and capacitance occur, with strong frequency dependence analyzed over 10^{-2} Hz to 10^{7} Hz. Similarly, polyimide/graphene oxide composites exhibit frequency-dependent dielectric constants that increase with filler content, interpreted through percolation threshold models.

The temperature dependence of frequency-dependent electrical properties, observed in chalcogenide thin films such as $Se_{80}S_{20}$, provides additional context for understanding the behavior of *$(Bi_{0,5}Sb_{0,5})_2Te_3$* films [24]. The shift in the minimum of temperature dependence according to $T_{\min} = \frac{R_{\text{int}}}{R_v^2}$ suggests that both crystallite and contact resistances exhibit distinct temperature dependencies, which could be exploited for temperature-compensated strain sensing applications.

While the present study provides valuable insights into the frequency-dependent tensoresistive behavior of *$(Bi_{0,5}Sb_{0,5})_2Te_3$* films, several limitations should be acknowledged. First, the measurements were conducted at room temperature; temperature-dependent studies would provide a more complete picture of the transport mechanisms. Second, direct microstructural characterization (e.g., scanning electron microscopy, transmission electron microscopy) of films before and after cyclic deformation would validate the proposed mechanisms of dielectric gap formation and contact degradation.

Future research should focus on optimizing film preparation parameters to achieve a balance between high tensoresistance sensitivity and mechanical durability. The development of composite structures incorporating flexible substrates and protective encapsulation layers, as demonstrated in recent flexible thermoelectric generators, could enhance the practical applicability of these materials [25]. Additionally, exploring the integration of $(Bi,Sb)_2Te_3$ films with advanced signal processing algorithms could enable more sophisticated fatigue damage detection and structural health monitoring capabilities.

Conclusion

This work confirms that composite ¿ thin films exhibit a strong frequency-dependent tensoresistive response that is significantly affected by cyclic mechanical deformation. Repeated loading up to 5×10^5 deformation cycles leads to a reduction of the critical frequency by nearly two orders of magnitude, which is associated with the formation of dielectric gaps at crystallite boundaries and the accumulation of mechanical fatigue damage. The observed frequency behavior is well described by an equivalent electrical circuit model incorporating crystallite resistance, inter-crystallite contact resistance, and shunting capacitance.

The high tensoresistive sensitivity and nonlinear deformation characteristics of the films are consistently interpreted within the framework of percolation theory, highlighting the strong influence of microstructural connectivity on electrical transport. The pronounced sensitivity of the electrical properties to cyclic loading suggests that these films are promising candidates for sensing accumulated fatigue damage in structural health monitoring systems, particularly for aerospace and other mechanically demanding applications.

Furthermore, the results are in good agreement with recent developments in flexible thermoelectric materials and strain-sensing technologies, emphasizing the multifunctional capabilities of Bi_2Te_3-based compounds. Optimization of fabrication parameters and integration into flexible device architectures may enable the development of self-powered monitoring systems that combine energy harvesting with real-time damage detection. Future studies should address temperature-dependent transport behavior, detailed microstructural characterization, and the fabrication of practical device prototypes to fully exploit the potential of ¿ films for next-generation flexible electronics and smart sensing applications.

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