Chaos in the transient current through As₂Te₃(In) and Mackey-Glass Simulation of Hysteresis Effect on glass substrates

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Abstract. $As_2Te_3(In)$ is a glass substrate (chalcogenide) that can be used in electronic devices in engineering such as computer memory arrays, display devices, optical mass memories because of its sensitivity to current transition. These glass substrates show chaotic behavior. In this study, we observed that by calculating the maximal Lyapunov exponents with non-linear time series analysis techniques. For this technique, at first linear auto-correlation function is used to show the delay time value in long- range correlation. However, the mutual information technique gives more reliable delay time value in short-range correlation. Estimated delay time values provide appropriate embedding dimensions by the false-nearest neighbors method. Attractor reconstruction is used for showing chaotic behavior, Hurst and Detrended Fluctuation Analysis (DFA) are used to support chaoticity. Hurst analysis describe the motion of transient current in time evolution. Calculated values of DFA give us long range power of correlation exponents. We observe slow transient effects causing hysteresis in the current-voltage characterization. We find that the current-voltage (IV) measurement delay time. Furthermore, As2Te3(In) shows hysteresis effect. A delay differential equation such as Mackey-Glass which has both periodic and aperiodic solutions can be a useful simulation tool for I-V characterization of glass substrate, which indicated chaos or non-periodicity in time series, is claimed to constitute a suitable model.

Keywords: Chalcogenides, Lyapunov Exponents, Correlation and delay time, Time Series Analysis, Hurst Analysis, Hurst exponent, Detrended Fluctuation Analysis, Hysteresis Effect, Mackey-Glass Equation

1 Introduction

The samples under investigation were set up as sandwiched metal-glass-metal structures with the glass as the isolating layer. 300 nm thick indium electrodes were thermally evaporated at 10^{-6} mbar on microscope glass slides cleaned in a solution. Afterwards, indium contacts were evaporated. The I-V measurements were performed by a programmable picoammeter/voltage source (Keithley, model 487) and a temperature controller (Lake Shore, model 300). The

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picoammeter and the temperature controller were interfaced to a PC through an interface card that automated data taking. The setup is schematically introduced in Fig. 1. The picoammeter model 478 utilized is equipped for reading currents in the range 10 fA to 2 mA. It additionally fills in as a DC voltage supply in the range up to 500V.



Fig. 1. Schematic of the experimental setup

The data of transient current against time (data points * 0.37s) at constant voltage of 2 V for $As_2Te_3(In)$ is presented as different sets with noise and without noise in Fig. 2. The noise was reduced from the raw data applying noise reduction tool on the TISEAN software package published by R. Hegger *et al.* [1] and Kantz and T. Schreiber [2]. The transient current in $As_2Te_3(In)$ glass films for times in the range of twelve thousand seconds has been analyzed. Several sets of data on different samples were taken at intervals of 0.37 s under 2 V at 23°C as room temperature (296 K).



Fig. 2. The current change in $As_2Te_3(In)$ under a constant applied voltage of 2V.

The data was originally taken to find a steady state value for the current in an experiment prepared to study mechanisms of conductivity in $As_2Te_3(In)$ thin film samples. The current was expected to settle down to a steady state value in a couple of hours. On the other hand, during this process, it was observed that the current against time (data points * 0.37s) plot shows a transient behavior characteristic of chaotic dynamical systems. At the point when an electric field

is connected to the thin films, the characteristic electron traps in films because of defects and impurities can offer ascent to the accompanying impacts: formation of new traps and annihilation of traps arising from the applied DC electric field. Subsequently, the electric field may be considered as an operator that may force the system, to damp and cause both impacts. However, the data looks more like the behavior of the transient current data for polymer thin films such as PMMA observed by A. Hacınlıyan *et al.* [3]. A characteristic of nonlinear systems related to chaos is forced damped motion. Another indicator of chaos was observed as non-periodicity in the signal. These characteristics steer us into analyze the transient current data using tools of nonlinear time series analysis methods.

In section 2 contains details of the analysis that has been performed and observations that have presented that the data show two different time scales and chaotic behavior have been seen in the transient data. In section 3 contains the Hurst (R/S) Analysis of $As_2Te_3(In)$ and results to obtain the motion of the current mechanism. In section 4 includes Detrended Fluctuation Analysis results of data sets. Section 5 includes simulation of Mackey-Glass equation for data sets which have hysteresis. In section 6 contains a brief discussion.

2 Chaoticity of the transient current in $As_2Te_3(In)$

Time series analysis is used for analyzing the data of $As_2Te_3(In)$ using TISEAN [1,2] software package. The formulas as part of the standard literature are used and applied. The reconstruction of the phase space from the scalar transient current s(k), where k means the kth time step, requires that we form vectors $\vec{y}(k)$ given by

$$\vec{y}(k) = [s(k), s(k+\tau), \dots, s(k+(m-1)\tau] \qquad \vec{y} \in \mathbb{R}^d$$
 (1)

where τ denotes the delay time and *m* denotes the embedding dimension. The scalar values s(t) are taken with respect to a sampling time, and the delay τ must be taken a multiple of it. If the time delay is too short the coordinates s(n) and $s(n + \tau)$ will not be sufficiently independent, hence the tangent jet cannot be sufficiently distinguishable. If the time delay is too large the correlation between s(n) and $s(n + \tau)$ will be lost. The meaningful time delay τ and the meaningful embedding dimension is found in order to construct time delay vectors and determine the number of parameters that correspond to the dimensionality of the system. The delay time is found by using Mutual Information (MUT) or correlation function (CORR). After determining the delay times, the embedding dimension is found by using the False Nearest Neighbors (FNN) method with using this appropriate values. The linear autocorrelation function determining the first zero for the determination of the time delay determined by Abarbanel [4] is given by

$$C_{1}(\tau) = \frac{\frac{1}{N} \sum_{m=1}^{N} [s(m+\tau) - \bar{s}] [s(m) - \bar{t}]}{\frac{1}{N} \sum_{m=1}^{N} [s(m) - \bar{s}]}$$

(2)

where $\bar{s} = \frac{1}{N} \sum_{m=1}^{N} s(m)$. The autocorrelation functions, which are the Fourier transform of the power spectrum - $C_1(\tau)$ versus τ , have first zeros at times of with respectively 5260 s (14197 time steps*0.37s) for the data set without noise and 5330 s (14351 time steps*0.37s) for the data set with noise.

Another method for the determination of the delay time is to find the first minimum of the average mutual information as a nonlinear correlation function given by Fraser and Swinney [5]

$$I(\tau) = I_{AB} = \sum_{a_i, b_j} P(s(n+\tau), s(n)) \log_2 \left[\frac{P(s(n), s(n+\tau))}{P(s(n+\tau))P(s(n))} \right]$$
(3)

 $P(s(n), s(n + \tau))$ is the joint probability at a time *n* measured s(n) later at a time $n + \tau$ measured $s(n + \tau)$. P(s(n)) is the probability of measured s(n) [2, 4]. The first minimum values are respectively at 20 s for the data set without noise and 35 s for the data set with noise. According this mutual information, the time delay has been chosen as the value of 20 s (54 time steps*0.37s without noise) and 35 s (93 time steps*0.37s - with noise). These values are more reliable estimations including nonlinear effects than linear autocorrelation time delay values. When the mutual information and the linear autocorrelation function are compared, two different time scales are observed: the signals forgets its state in between 20 s and 35 s when nonlinearity is considered, but they seem to be linearly correlated for up to 5260 s and 5330 s. This manner implies that there may be two different time mechanisms which go to two timing scales. The second step in phase space reconstruction is choosing the embedding dimension by the method of false nearest neighbors [4]. The fraction of false neighbors is presented against the embedding dimensions for different delay times respectively 20 s - 35 s (time delays for mutual information) 5260 s -5330 s (time delays for autocorrelation function). The ratio of false neighbors drops drastically after embedding dimension four for a delay time of 20 s and 35 s; hence an embedding dimension of four is a good estimate for the minimal embedding dimension.

From minimum to larger values of embedding dimension is represented by scaling considerations in the Lyapunov exponent calculation. For detecting the presence of chaotic behavior in the transient data, the maximal Lyapunov exponent is calculated by

$$S(\Delta n) = \frac{1}{N} \sum_{n_0}^{N} \ln\left(\frac{1}{|u_n(\overline{s_0})|} \sum_{\overline{s_n} \in u(\overline{s_{n_0}})} \left| \overline{s_{n_0 + \Delta n}} - \overline{s_{n + \Delta n}} \right|\right) \tag{4}$$

 \vec{s}_{n_0} is the embedding vector as a reference point. All neighbors are selected with distance smaller than the ε value (denoted as $u_n(\vec{s}_0)$) and are averaged over the distances of them to the reference point at time Δn . The distances go as $e^{\lambda \Delta n}$, where λ is the maximal Lyapunov exponent. If $S(\Delta n)$ shows a linear robust increase for Δn then the slope is taken as maximal Lyapunov exponent described detailly by Kantz [6]. The slope values of the persistent linear increases give the Lyapunov exponent values. Table 1 and Table 2 shows respectively these Lyapunov exponent values for different embedding dimensions.

m=2	m=3	m=4	m=5
0.0095	0.0105	0.0113	0.0119

 Table 1. Lyapunov exponent values for different embedding dimensions of data set with noise

m=2	m=3	m=4	m=5
0.0102	0.0112	0.0120	0.0125

 Table 2. Lyapunov exponent values for different embedding dimensions of data set without noise

From Table 1 and Table 2 values, the average values of $0.0108 \pm 0.0004 \ s^{-1}$ for data set have noise and $0.0115 \pm 0.0004 \ s^{-1}$ for data set have not noise are estimated for the maximal Lyapunov exponent. These positive values getting from the slopes indicating the time evolution of the transient current has chaotic behavior.

3 Hurst (**R**/**S**) Analysis of $As_2Te_3(In)$

The Hurst exponent is obtained utilizing the standard approach and it is a numerical approach to deal with the consistency of a period arrangement in a time series. If the Hurst exponent (H) is near 0.5, the process is a random walk well known as Brownian motion. A Hurst example (H) in the range 0 < H < 0.5 suggests non-random behavior in a time series. A Hurst exponent (H) in the range 0.5 < H < 1 suggests a time series with long range, persistent advancement. In Fig. 3, the fitting functions are applied to the Hurst Analysis to get Hurst exponents. The data sets have three regimes as seen from graphs. Table 3 gives the results of these regimes to interpret the behavior mechanism of transient current in time evolution.



Fig. 3. Fitting graphs for evaluating the Hurst exponent values

	Hurst exponents for data set with noise	Hurst exponents for data set without noise
First regime	0.2805	0.3016
Second regime	0.0924	0.0840
Third regime	0.3408	0.3364

Table 3. Hurst exponents of the fitting graphs

4 Detrended Fluctuation Analysis (DFA) of As₂Te₃(In)

Detrended Fluctuation Analysis is used on the data sets to sustain the indication of changing regimes. Detrended Fluctuation Analysis (DFA) is a scaling analysis technique used to determine long range power law correlation exponents expressed by C. K. Peng *et al.* [7, 8]. It is obtained by integrating the time series of length N, then dividing the outcome into boxes of equivalent length, n. A least squares line is fit to the data in each box of length n. The y coordinate of the straight-line sections is signified by $y_n(k)$. Next, these integrated time series, y(k), is detrended by subtracting the local pattern, $y_n(k)$, in each box. The root-mean-square fluctuation of this integrated and detrended time series is determined by

$$F(n) = \sqrt{\frac{1}{N} \sum_{k=1}^{N} [y(k) - y_n(k)]^2}$$
(5)

This calculation is rehearsed over all time scales (box sizes) to describe the relation between F(n), the average fluctuation, as a routine of box size, n. A straight relation on a log-log plot demonstrates the nearness of power law scaling. Under such conditions, the fluctuations can be identified by a scaling exponent, α , with that $F(n) : n^{\alpha}$. A crossover in the scaling exponent, α , shows a transition from one kind to a different kind of basic relationship, because of a transition in the dynamic properties described by K. Hu *et al.* [9] and Z. Chen *et al.* [10]. In Fig. 4, the fitting functions are applied to the DFA. The data sets have two regimes as seen from graphs. Table 4 gives the results of these regimes to interpret the long-range power law correlation exponents.



Fig. 4. Fitting graphs for evaluating the correlation exponent values

	Correlation	Correlation
	exponents for data	exponents for data
	set with noise	set without noise
First regime	1.4526	1.5890
Second regime	0.8142	0.8101

Table 4. Correlation exponents of the fitting graphs

5 Hysteresis Effect of $As_2Te_3(In)$ and Simulation of Mackey-Glass equation

The data sets which have hysteresis shown in Fig 5. have been obtained as the current values versus the changing voltage (0 V – 4 V) values in constant interval of ΔV at varying time steps of Δt . Respectively, the program has been arranged as forward bias from 0 V to 4 V and backward bias from 4 V and 0 V. So, the hysteresis and memory effect on semiconductor glass substrates

(chalcogenides) of $As_2Te_3(In)$ has been broadly examined. The *Mackey-Glass* equation has applied to our empirical data set of $As_2Te_3(In)$ glass substrate. The artificial data set is simulated to determine the chaotic mechanism of the empirical data set with using a tool of MATLAB named as NAR (nonlinear autoregressive) Neural Network Analysis applied by P. Potocnik [13]. The *Mackey-Glass equation* is the nonlinear time delay differential equation

$$\frac{dx}{dt} = \beta \frac{x_{\tau}}{1 + x_{\tau}^n} - \gamma x \quad \text{where} \quad \gamma, \beta, n > 0.$$
(6)

where β , γ , τ , n are real numbers and x_{τ} represents the value of the variable x at time $(t - \tau)$ [14]. Depending on the values of the parameters, this equation displays a range of periodic and chaotic dynamics. Fig. 7 shows us the dynamics in the *Mackey-Glass equation* which based on the parameters for $\gamma = 0.53$, $\beta = 1.02$, $\tau = 30$, n = 7.



Fig. 5. Data set which has hysteresis (I versus V at varying time)



Fig. 6. Dynamics in a piece of empirical data set of $As_2Te_3(In)$ (500 samples arbitrarily chosen from entire data set)



Fig. 7. Dynamics in simulation of *Mackey-Glass equation*, for $\gamma = 0.53$, $\beta = 1.02$, $\tau = 30$, n = 7

The relation between the validation data and training data sets which has nearly close prediction depends on the error. In these graphs, the simulations have taken first 300 numbers of samples from a piece of the empirical data set to recognize the initial dynamics of the transient current mechanism. After this process, the simulations have iterated the last 200 numbers of samples to validate artificial data. When the numerical difference between sampling data and predicted data has been calculated, the error has been determined. Fig. 8 shows how dynamics of dataset have changed.



Fig. 8. Dynamical graph of the simulation

In Fig. 8, simulated data had best validation performance when the number of iterations has increased. Meaningfully, more accurate results have been obtained when simulation codes have been mostly iterated. Furthermore in Table 5 and 6, the maximal Lyapunov exponents for validation and prediction values of the simulation have been calculated. So, they could be compared for the simulated dataset and empirical dataset.

m=2	m=3	m=4	m=5
0.0133	0.0159	0.0160	0.0178

 Table 5. Lyapunov exponent values for different embedding dimensions of data set for validation

m=2	m=3	m=4	m=5
0.0117	0.0181	0.0194	0.0191

 Table 6. Lyapunov exponent values for different embedding dimensions of data set for prediction

6 Conclusions

The transient current in $As_2Te_3(In)$ shows chaotic behavior with a positive Lyapunov exponents as $0.0108 \pm 0.0004 \ s^{-1}$ for data set with noise unfiltered and 0.0115 \pm 0.0004 s⁻¹ for data set without noise, and an embedding dimension of four. The Lyapunov exponent values for different embedding dimensions are very close to each other, so it can be said that the data which has noise and is purified from noise point to analogous transient current mechanism at this environmental condition. Both the false nearest neighbors method and application of maximal exponent were used as different approaches to gain reliable value of the embedding dimension. The complex identity of glass substrates implies many degrees of freedom and a multifractal structure. So, obtaining identical results under same conditions will not be so easy. There are additional important studies about using of nonlinear methods to analyze the conduction mechanism in different amorphous structures which have irregular behavior. [3,11,12,15] The first zero of the linear autocorrelation function and the first minimum of the mutual information function give different time scales. There are local minima values in the mutual information function, but these values have same order of magnitude under the first zero of the autocorrelation function. Consequently, this indicates two different time scales in the conduction mechanism of glass substrates. When the electric field is applied, the system is forced because of created new traps. Also, it behaves a dissipative agent by annihilating them. Many conflicting manners exhibit two significances, so these significances may cause a bifurcation effect and may be have two different time scales. If both occur in a complex structure of a glass substrate, the chaotic time evolution of the current appears. Accordingly, the Hurst Analysis shows that the data sets have three regimes and three different Hurst exponent values. All of them are less than the critical value of 0.5 for Hurst Analysis. This implies that there is non-random behavior mechanism of transient current of time evolution for the data sets. Only second regime has smaller value than others. It can be considered, there is a transition that the fluctuations of the current through glass substrate saturate between this gap belongs to the second regime. After that, the conduction mechanism of the sample recognizes the condition and it properly behaves related to its memory. On the other hand, two different regimes observed on Detrended Fluctuation Analysis. The values of the slopes are 1.4526 and 0.8142 for the data with noise, and 1.5890 and 0.8101 for the data without noise. The observed slope points indicate a change in correlation properties that sustain the change in the dynamics of the system observed during the above investigation of maximal Lyapunov exponents. Definitely, distinction of the slopes of the data without noise is more than the distinction observed in the data with noise. The range of maximal Lyapunov exponents for the data without noise $(0.0115 \pm 0.0004 \text{ s}^{-1})$ is greater than the maximal Lyapunov exponents for the data with noise (0.0108 $\pm 0.0004 \ s^{-1}$). So, the change observed in the maximal Lyapunov exponents of two types of data sets is additionally identified by DFA. The fact that the rescaled range analysis gives a steeper slope followed by a less steep slope is compatible with the DFA results. The fact that the rescaled range analysis reverts to the higher slope is probably an artifact of its sensitivity to extreme values. By applying the simulation of the Mackey-Glass equation with using computational environment, artificial data has been compared with empirical data. As final concept, the physical behavior of the samples has been definitely explained as a general phenomenon. The range of maximal Lyapunov exponents for prediction dataset $(0.0171 \pm 0.0007 \text{ s}^{-1})$ is greater than the range of maximal Lyapunov exponents for validation dataset (0.0157 \pm 0.0007 s⁻¹). The maximal Lyapunov exponents of validation and prediction dataset have similar values with the maximal Lyapunov exponents of empirical dataset. This implies that the Mackey-Glass equation is a suitable simulation method to analyze and predict transient current mechanism and hysteresis of $As_2Te_3(In)$ glass substrates.

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