Quasiperiodic and chaotic self-excited voltage oscillations in $TlInTe_2$

M. P. Hanias, J. A. Kalomiros, Ch. Karakotsou, A. N. Anagnostopoulos, and J. Spyridelis
Solid State Section 313-1, Physics Department, University of Thessaloniki, 54006 Thessaloniki, Greece
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The electrical behavior of the $TlInTe_2$ ternary semiconductor is studied in the negative-differential-resistance region of its S-type I-V characteristic. Self-excited voltage oscillations in this region, with an amplitude of about 5 V were monitored at temperatures above 70 K. An analysis of the dynamic behavior of these oscillations by means of the Grassberger and Proccacia method shows the existence of two components in the signal, a quasiperiodic component and a chaotic component. Furthermore, this analysis leads to the estimation of fractal dimensions, minimum embedding dimensions and Kolmogorov entropies that characterize the two components.

I. INTRODUCTION

$TlInTe_2$ is a p-type semiconductor with a fibrous structure. $In^{3+}Te_2^{2-}$ groups form chains extending along the c axis of the material. These negatively charged chains are bonded together by Tl^+ ions. The resulting tetragonal lattice is characterized by group symmetry D_2^{18}-$I4/mcm$.[1,2]

As already reported in a previous paper,[3] $TlInTe_2$ crystals present interesting nonlinear electrical behavior, such as, switching effects and a negative-differential-resistance (NDR) region in their S-type I-V characteristics. In a preliminary work,[4] we studied separately the Ohmic and NDR region. We used Arrhenious diagrams to evaluate the parameters of the acceptor levels that regulate the electrical conductivity in the linear region. The energy positions of the acceptor levels were found to be at 0.41 and 0.030 eV above the top of the valence band with concentrations 4×10^{14} and 1×10^{19} cm^{-3}, respectively.

To interpret the current-controlled negative-differential-resistance (CCNDR) region that appears at moderate and high current densities, a dominating electrothermal process was taken into account. The effect that strongly corroborates this interpretation is the strong elevation of the temperature of the sample and its distribution along the I-V characteristics. The electrothermal model used to describe qualitatively and quantitatively these data can well account both for the absolute values of the temperature of the sample for each current value, as well as for the dependence of the threshold voltage V_{th}, at which the initiation of the NDR region occurs, on the ambient temperature.[4]

In the NDR region of these characteristics, self-excited oscillations of the voltage were also observed.[5] In the present paper, we report on the dynamic behavior of these oscillations. Our purpose is to establish the principally chaotic nature of the effects that dominate the NDR region of the I-V characteristic of $TlInTe_2$ single crystals. We also present a quantitative analysis and produce values for characteristic parameters that govern the chaotic effects in this semiconductor system. The results will be discussed in the context of effects observed in similar and other semiconductor materials.

II. EXPERIMENTAL DETAILS

Semiconducting $TlInTe_2$ samples used in our electrical measurements were cleaved from crystals grown by direct melting of pure stoichiometric amounts of their constituent elements.

The samples were rectangularly shaped with parallel faces and dimensions of the order $10\times1\times1$ mm^3. In, Au, and Ag were found to form Ohmic contacts of low resistance with $TlInTe_2$. This was proved by the four-contact method, by successive resistivity measurements on samples of different thicknesses, as well as by photoconductivity measurements. In the present work, evaporated In stripes were used as contacts. The current-providing contacts were applied on the ends of the rectangular samples and oriented so that the current flowed along the c axis of the material. A current source (model 225 by Keithley Instruments) was used to control the current in the measurement of the S-type I-V characteristics. The corresponding voltage drop was registered and measured by a Keithley voltmeter (610A). Low-frequency voltage oscillations that appear in the NDR region of the I-V characteristics were monitored by an X-Y recorder. In the high-frequency range a storage oscilloscope was used. The samples were mounted on the copper cold finger of a liquid-helium cryostat that enabled electrical measurements down to 10 K. An appropriate feedthrough was used to allow the electrical wiring inside the cryostat. A copper cup was used to minimize thermal losses and to electrically shield the specimens. All measurements were performed in a vacuum below 10^{-5} Torr. Coaxial cables were used in the circuitry in order to avoid the influence of external noise. With all these precautions noise was restricted to a level below 0.1%.

III. ANALYSIS OF THE VOLTAGE OSCILLATIONS

A. Characterization of the attractor

In Fig. 1, a representative current-voltage characteristic is shown, registered on a sample of $TlInTe_2$, at an ambient temperature of 77 K. Voltage oscillations were ob-

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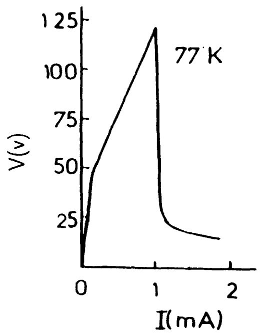

FIG. 1. A representative current-voltage (I-V) characteristic as registered on $TlInTe_2$, at an ambient temperature 77 K.

served in the NDR region, both just after exceeding the threshold value, I_{th}, as well as at higher values. I_{th} is the current value that corresponds to the initiation of the NDR region. In the region of oscillations, voltage values of the I-V curve were obtained by averaging over long-time intervals. A representative example of such a signal is shown in Fig. 2(a). This wave form was monitored for a current value $I = 1.15$ mA, in the NDR region of the I-V characteristic shown in Fig. 1. Figures 2(b) and 2(c) show the enlargement of two different regions a, b of the signal of Fig. 2(a). In Fig. 3, the power spectrum of the whole signal shown in Fig. 2(a) is presented. Similar results were obtained from the time series of Figs. 2(b) and 2(c). The spectrum of Fig. 3 consists of pronounced frequency lines, like those indicated by arrows and a broadband background, where the lack of pronounced frequencies is evident. This broadband background is not due to white noise, which lies at a level lower than 0.01 V, in Fig. 3, as already mentioned in Sec. II. The form of the power spectrum is indicative of the nonperiodic nature of the signal of Fig. 2(a). This is consistent with the behavior of the nonperiodic trajectories in the phase portrait of Fig. 4. They seem to lie within the basin of a seemingly uniform attractor. Phase portraits obtained for the signals of Figs. 2(b) and 2(c) are very similar to that of Fig. 4.

Qualitatively, the phase portrait of Fig. 4 can be considered as consisting of rather quasiperiodic components, highly dispersed by other, chaotic components. For a quantitative study of the signal of Fig. 2(a), we applied the well-known method, proposed by Grassberger and Procaccia.[6,7] According to Takens theorem,[8] we used the digitized form of the signal shown in Fig. 2(a), in order to reconstruct the original phase space, where the attractor characterizing the system is properly embedded. We calculated the correlation integral $C_m(2,l)$, which is defined by the following formula:[9]

$$\sum_i p_i^2 = C_m(2,l) = \lim_{N\to\infty} (1/N^2) \times \sum_{i,j=1}^{N} \Theta\left\{ l - \left[\sum_{k=1}^{m} |X_{j+k} - X_{i+k}|^2 \right]^{1/2} \right\} .$$

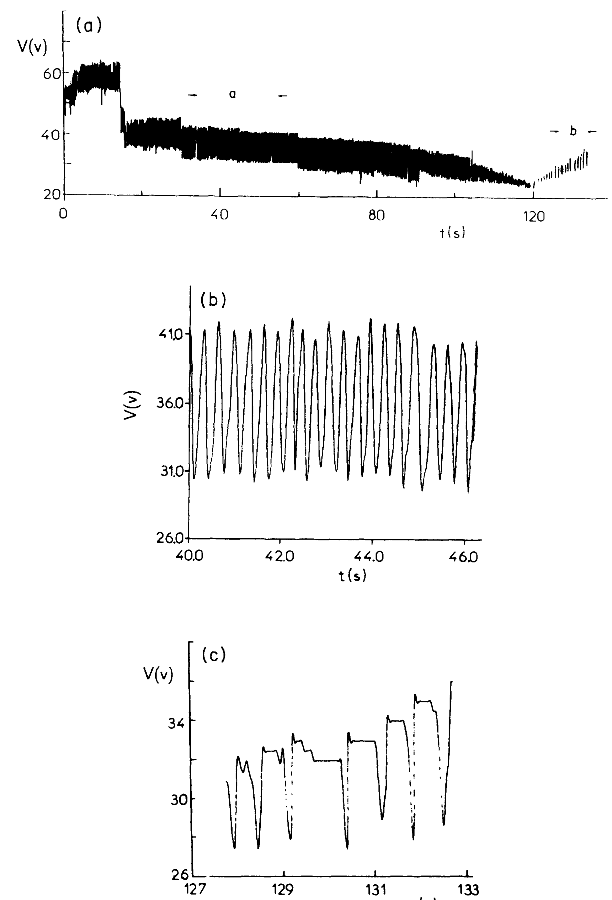

FIG. 2. (a) Typical voltage oscillations as monitored in the NDR region of the curve presented in Fig. 1, for a fixed current value $I = 1.15$ mA. (b) and (c) Enlargements of the regions a and b of the signal in (a).

In the above equation, N is the number of the experimental points, X_i is a point in the m-dimensional phase space with $X_i = \{U(t_i), U(t_i+\tau), \ldots, U[t_i+(m-1)\tau]\}$, τ is the time delay parameter, and Θ is a step function (Heaviside function) that simply counts the points in the phase space with mutual distances less than l. The proper value of τ can be chosen using the autocorrelation function of the measured time series.[10,11] The numerical determination of the correlation integral is based on the box count-

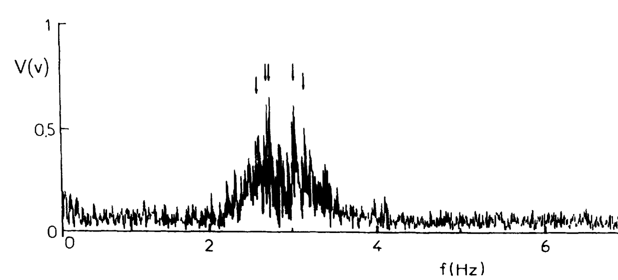

FIG. 3. Power spectrum of the total signal shown in Fig. 2(a). Power spectra of the signals of Figs. 2(b) and 2(c) look very similar to it.

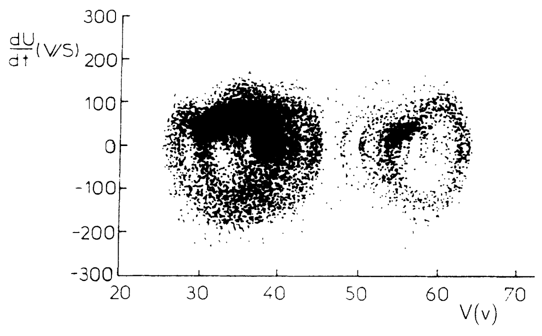

FIG. 4. The phase portrait of the total signal, shown in Fig. 2(a).

ing method. Applying the above expression for $\tau=1$ (in units of sampling rate), we have calculated the correlation integral as a function of l for different embedding dimensions m, Fig. 5. The slope of the linear parts of these double-logarithmic plots provides information about the nature of the attractor.

As it is evident from the curves of Fig. 5, their lower regions consist of two linear parts, with quite different slopes ν_1 and ν_2. These parts become parallel and equispaced at higher m values. In the first group of these linear segments, which appear at intermediate l values, the correlation integral is proportional to l^{ν_1}. ν_1 approaches the asymptotic value 1.08, as $m \rightarrow \infty$, Fig. 6(a).

On the other hand, in the second group of the linear segments, which appears at low l values, the correlation integral is proportional to l^{ν_2}. From Fig. 6(b) it is obvious that ν_2 is not proportional to m (i.e., the signal is not a random one), but seems to saturate at the value 2.86. This value is much higher than ν_1 and confirms the different behavior of the signal component, from which this part arises.

One has to point out that ν_1 saturates already at low m values, i.e., ν_1 already approaches its asymptotic value by

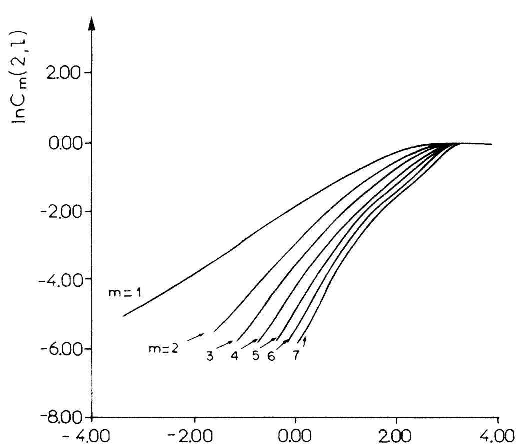

FIG. 5. The correlation integral $C_m(2,l)$ vs l, with the embedding dimension m as a parameter, obtained with a delay time $\tau=1$ (units of sampling rate).

80%, for $m=2$ [Fig. 6(a)]. This means, that this component needs only a two-dimensional phase space for its appropriate embedding, as expected. On the contrary, ν_2 seems to saturate at higher m values. In this case, this component needs a high-dimensional phase space for its appropriate embedding, as expected.

The Grassberger and Procaccia method leads to the estimation of the Kolmogorov entropy K_2, a measure of the information loss of the system. $C(l)$ scales with K_2 according to an exponential law[12] of the form $C(l) \sim e^{-m\tau K_2}$. With increasing m, K_2 decreases monotonously, tending to zero in the case of the component with slope ν_1. This behavior is shown in Fig. 7 (curve a) and is evidence for a quasiperiodic signal component. In the case of the component with slope ν_2, K_2 tends to the asymptotic value 1 bits/τ (Fig. 7, curve b), which is positive and finite, a consistent evidence for the existence of a chaotic attractor, governing the motion of this component in the phase space.

B. Introduction of generalized dimensions

The method of Grassberger and Procaccia, as presented in the previous paragraph, provides reliable results, only in the case of rather uniform attractors. To check

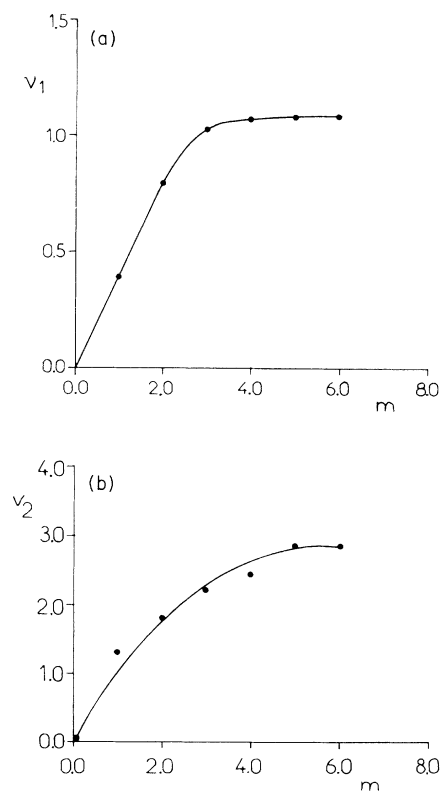

FIG. 6. (a) The slope ν_1 of the upper linear parts of the curves in Fig. 5 vs the embedding dimension m. For high values of m the curve saturates at the value $\nu=1.08$. (b) The slope ν_2 of the lower linear parts of the curves in Fig. 5 vs the embedding dimension m. Note that with increasing m, the curve clearly deviates from the proportionality to m, tending to an asymptotic value higher than 2.86.

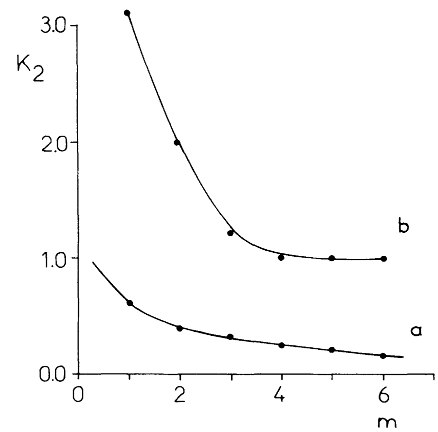

FIG. 7. The K_2 entropy plotted as a function of m for the upper and lower linear segments of the curves of Fig. 5, curves a and b, respectively. (Curve *a*) For high values of m this curve tends to $K_2=0$. (Curve *b*) For high values of m this curve tends to $K_2=1$ bits/τ.

the homogeneity of the attractor we have introduced the generalized dimensions D_q.[13] The generalized dimensions D_q are related to the generalized correlation integral $C_q(l)$ through the expression

$$D_q=-\lim_{l\to 0}[1/(q-1)][\ln C_q(l)/\ln l] ,$$

where

$$C_q(m,l)=\sum_i p_i^q ,$$

The correlation integral $C_q(l)$, measures the probability of finding q points of the attractor in the m-dimensional phase space, at a distance smaller than l.[14,15] The D_q's measure correlations between different points on the attractor and are, therefore, useful in characterizing its inhomogeneous static structure. For $q=0$, we obtain D_0, which is the ordinary fractal dimension of the attractor (Hausdorff dimension), for $q=1$ we obtain the information dimension D_1 and for $q=2$ the correlation dimension D_2.[16] Using the last two relations we have calculated D_0 and D_1. These values are very close to D_2, as it was calculated applying the Grassberger-Procaccia method, a fact providing evidence that the attractor can be considered to be approximately homogeneous for these q values.

IV. DISCUSSION

In the present paper, we studied the nonlinear behavior of $TlInTe_2$ semiconductor crystals and evaluated the observed nonperiodic oscillations by means of a well-established procedure. The obtained results strongly support the idea that, the initial experimental signal consists simultaneously of a quasiperiodic and a chaotic component.

The two components, quasiperiodic and chaotic, are present throughout the whole NDR region, i.e., for every value of the control parameter I. Changing the current I, the contribution of each component in the total signal varies. At low current values the signal is dominated by the quasiperiodic contribution, while at higher values, the chaotic component is dominant.

In the literature, double-logarithmic plots of $C_m(2,l)$ as a function of l showing two distinct linear parts, are usually correlated with the simultaneous existence of two signals, one of deterministic chaotic origin and one due to external white noise. In such cases,[17–19] the upper linear part comes from the chaotic signal, while the lower one is due to white noise. The main feature of the part arising from the noise is that its slope does not tend to an asymptotic finite value with increasing m, but it also increases, tending to infinity. Such a part with slope monotonously increasing with m is not present in the curves of Fig. 5, corroborating that the noise level present in the measurements was low. Thus, a decisive factor for the classification of the linear parts of the curves of Fig. 5, was that their slopes tend to finite asymptotic values. Similar results, i.e., double logarithmic plots of $C_m(2,l)$ as a function of l consisting of two distinct linear parts with different slopes are reported also for the system studied in Ref. 20. According to Ruelle[21] this is expected for a system with two noninteracting attractors of different amplitudes.

To the best knowledge of the present authors this is one of very few papers on the appearance of self-excited oscillations in a bulk semiconductor at temperatures higher than 10 K. It should be stressed that the crystal presented in this work shows the same, though less pronounced, dynamic behavior up to room temperature. Reports on the appearance of chaotic effects at temperatures lower than 10 K can be found in Refs. 22 and 23. These authors present dynamic behavior observed in Ge and GaAs semiconductors. The mechanism responsible for their effects is impact ionization, triggered at very low temperatures.

On the other hand, high temperature, self-excited dynamic behavior, similar to the one reported in the present paper, has also been observed in the related chainlike semiconductor $TlGaTe_2$.[24] In general, electrothermal current filamentation can account for the dynamics observed in this later kind of semiconductor materials.[24,25]

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