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Switching and memory effects in partly crystallized amorphous $Ge_2Sb_2Te_5$ films in a current controlled mode

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ABSTRACT

Switching and memory effects in as-deposited amorphous $Ge_2Sb_2Te_5$ films with a considerable concentration of crystalline nuclei have been investigated. Variation of the phase composition of the sample has been confirmed by Raman spectroscopy data. The influence of nuclei on parameters of the current voltage characteristic has been studied. Significant variation of initial resistance and threshold voltage due to a different nuclei configuration has been observed. In some cases the current voltage characteristic was monotonous i.e. the intrinsic S-shape of the current–voltage characteristic disappeared and memory recording occurred without prior switching. The measurements were made in a current controlled mode which allowed conducting a thorough investigation of switching and current filament formation.

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1. Introduction

Recent progress in optical and electrical information recording on chalcogenide glassy semiconductors (CGSs) [1–4] has attracted considerable interest to unusual electrical properties of these materials [5]. First, chalcogenide glasses contain a high concentration of intrinsic defects with negative correlation energy, also called negative-U centers. These defects are responsible for the Fermi energy pinning near the middle of the band gap [6,7]. Secondly, the resistivity of CGSs exponentially decreases with increasing voltage [8]. In strong electric fields an electrical breakdown occurs and the CGS switches from a highly resistive to a conductive state. The switching effect is reversible and the CGS recovers the highly-resistive state after the voltage is removed [9–11].

In the conductive state the heat generation is large; therefore the active volume heats up and crystallizes. So CGSs can store the low-resistive state; therefore this effect is called memory effect [11]. The memory effect in CGSs is utilized in the phase-change random access memory (PRAM), a promising non-volatile memory technology which allows considerably faster read and write speed than flash memory [2].

When switching the resistivity of CGSs decreases by several orders of magnitude and in the conductive state of the amorphous phase it is close to the resistivity of the crystalline phase [12]. This allows using voltage pulses with a close height to crystallize (set) and amorphize (reset) the active region. Therefore the switching effect makes CGSs an ideal material for PRAM.

Though the switching effect in CGS was first observed in the early 60's, the physics of this effect remains unclear. It is believed that in micron-thick samples (>5 μm) the switching is due to a thermal breakdown [13]. Meanwhile in thin-film samples (<1 μm) the threshold electric field is sufficiently higher. Therefore it is widely accepted that electronic processes play an important role. From one point of view the switching effect is purely electronic [14–17] and from another point of view thermal processes still play an important role [18–22]. In the latter case the influence of the Joule heating is important along with electronic processes. At the present time it is obvious that understanding of the physical mechanism of the switching effect will help to improve the characteristics of the PRAM elements.

Usually the switching and memory effects are studied in homogeneous amorphous samples. However the aim of the present paper is to investigate switching and memory effects in partly crystallized inhomogeneous samples with a considerable concentration of crystalline nuclei. We have measured current–voltage characteristics (CVCs) of different points of the same sample. This allowed us to investigate the influence of crystalline inclusions at switching and memory effects and at the same time eliminated the influence of other factors, such as details of film preparation technology.

2. Experimental

$Ge_2Sb_2Te_5$ alloy was prepared by the quenching technique, and materials (99.999%) were weighed according to their atomic percentage and then sealed in quartz ampoule in a vacuum of $5\cdot10^{-3}$ Pa. The ampoule was rocked frequently for 6 h at a maximum temperature

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750 °C to make the melt homogeneous. Then the melt was quenched in a switched off furnace.

50 and 500 nm thick $Ge_2Sb_2Te_5$ films were deposited by the flash thermal evaporation of synthesized material on Si (100) substrates coated with a conductive Au or ITO layer which was used as a bottom electrode. The basic chamber pressure was 10^{-4} Pa, the temperature of the evaporator was 630 °C and the temperature of substrates was 50 °C. The deposition rate was 3 nm/s.

The composition of the thin-film samples was measured using an electron probe microanalyzer (EPMA) Camebax (Cameca). The analysis was made for five different points of the sample. A qualitative EPMA analysis was used to identify the elemental composition. Then the concentration of each element was determined by comparing the intensity of analytic lines from the sample with corresponding standard specimens.

Variations of the phase composition in as-deposited samples were identified using Raman spectroscopy. Measurements were made using a Horiba Jobin-Yvon T64000 spectrometer. Second harmonic of a Nd: YAG laser (532 nm) was used for excitation; laser beam was focused to a 5 μm spot with an intensity of 500 W/cm^2. Doubling of the laser intensity did not result in a qualitative change of the Raman spectra; this indicates that no heating of the sample occurs while measuring.

Additionally the structure of the sample surface has been investigated with a scanning electron microscope (SEM) Quanta3D 200i. Measurements were carried out in a high vacuum mode using a 30 kV accelerating voltage; the resolution was equal to 3.5 nm.

Current–voltage characteristics of different points on the sample surface were measured using a golden pressed contact mounted on a two-axis stage. The area of the sample was approximately 1 cm^2 and the distance between the points was approximately 1 mm. The area of the contact was approximately 10^{-4} cm^2.

The measurements were made in a current controlled mode. Triangular current pulses were formed by an 8-bit digital to analog converter (DAC). Current was increased stepwise; the duration of each step was equal to 4 μs, therefore the length of the whole pulse was approximately equal to 2 ms. Voltage on a sample was measured using a digital oscilloscope.

First we have measured the initial resistance of the point using a current pulse with $I_{max}=6.5$ μA. The corresponding value of the measured voltage was a few tenth of a volt and therefore does not switch the sample. Then switching and memory effects were investigated using a current pulse with $I_{max}=8.2$ mA. Typical oscillogram of the applied current and sample voltage is shown in Fig. 1. The threshold switching voltage U_{th} is the maximum voltage in the highly-resistive state and the holding voltage U_h is the voltage during the formation of the memory state.

Usually CVCs of chalcogenide glasses with switching are measured in a voltage controlled mode. Voltage pulses of a given shape are applied to the sample and a load resistor connected in serial. The voltage on the load resistor is measured to calculate the current through the sample; therefore the load resistor is also called a measuring resistor. When switching the sample resistance decreases and the applied voltage remains constant. Therefore the current through the sample increases abruptly [23]. High measuring resistors can be used in order to decrease the current jump; however this would limit the maximum current at a given voltage and decrease the accuracy of voltage measurement.

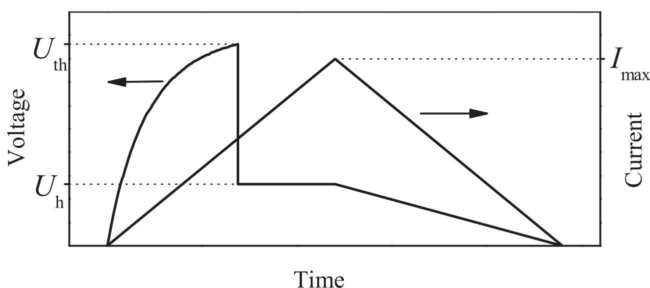

Fig. 1. Typical shape of observed oscillograms.

In the current controlled mode the voltage is measured as a voltage drop across the sample. When switching the sample resistance decreases abruptly; therefore the voltage decreases while the current remains constant. The latter makes possible to observe the filament formation process. Therefore the suggested measuring technique is preferable.

The main error was introduced by the digital oscilloscope and was equal to 1% of the measurement range. This means that the absolute error was 0.03 V for measurements at low current and 0.1 V for measurements of the switching effect. Also we have to note that the top electrode contact area may be different at different points. Our estimations show that the variation of the contact area is less than 10%.

3. Results

The quantitative EPMA analysis indicated that the composition of samples is $Ge_2Sb_2Te_5$ and a deviation of the concentration of each element does not exceed 2–3%.

The Raman spectra of all points of the sample have a typical for amorphous $Ge_2Sb_2Te_5$ double-humped shape in the region 100–170 cm^{-1} [24–26]. In Fig. 2 we represent three Raman spectra measured at randomly selected points on a sample surface within an area ~1 cm^2. Decomposition of the experimental data into two Gaussian curves indicates that positions of the band maxima are approximately the same; however, the integrated intensities of each band I_1 and I_2 vary at different points of the sample. The ratio of I_2 to I_1 was varied mainly between 0.4 and 0.6, however in several points near the sample edge the ratio was approximately 0.2. The high-frequency band (I_1) with a maximum in the region 150–160 cm^{-1} mainly corresponds to Sb_2Te_3 oscillations [27]. Oscillations of the $GeTe_4$ tetrahedron form the low-frequency shoulder (I_2) around 125–130 cm^{-1} [25,28] and also make a small contribution to the high-frequency band [29].

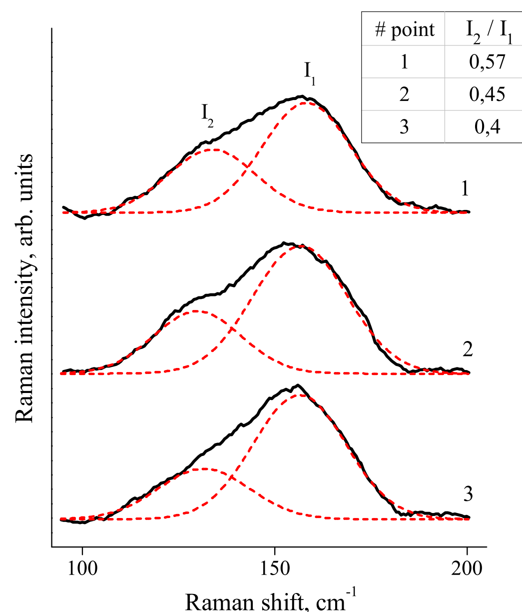

Fig. 2. Raman spectra of three randomly selected points of $Ge_2Sb_2Te_5$ thin film. Red dashed lines show the decomposition of the Raman spectra into two Gaussian curves. The band intensity ratio is shown in the table.

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