Study of the charge transport mechanism in pulsed laser deposited AlN:Cr films

I P Minkov¹,², S Simeonov¹, A Szekeres¹, C Ristoscu³, G Socol³, S Grigorescu³, I N Mihailescu³

¹Georgi Nadjakov Institute of Solid State Physics, Bulgarian Academy of Sciences, 72 Tsarigradsko Shausse, 1784 Sofia Bulgaria
²Technical University – Sofia, 8 Kliment Ochridski blvd, 1000 Sofia, Bulgaria
³National Institute for Lasers, Plasma and Radiation Physics, PO Box MG-54, Ro-77125, Bucharest – Magurele, Romania

E-mail: ipmincov@tu-sofia.bg

Abstract. AlN thin films doped with Cr (AlN:Cr) were prepared on p-Si(100) substrates by pulsed laser deposition. Al–AlN:Cr–Si MIS structures were formed and their current–voltage and 1 MHz admittance characteristics were measured and analyzed. The current through the MIS structure is proportional to the square of the applied voltage, which is evidence that the current is limited by space charge at traps in the AlN:Cr films. These traps are responsible also for the observed negative differential resistance and hysteresis of the current–voltage characteristics. The C–V and G–V characteristics measured at 1 MHz and the impedance measurements in 500 Hz–500 kHz frequency range without applying bias voltage are also influenced by the traps in the AlN:Cr films.

1. Introduction

The possibility for applying diluted magnetic semiconductors (DMS) in spintronics devices and structures has led to preparation and investigation of different DMS thin films. DMS is formed when metal ions are replaced by magnetic ions of transition metals such as Co, Cr or Mn. Ferromagnetism with Curie temperatures as high as 900°C is observed in Cr-doped AlN (denoted further as AlN:Cr) films [1]. For preparation of these films, the incorporation of Cr ions is accomplished usually by Cr co-evaporation during AlN MBE deposition [2] or by Cr ion implantation into already deposited AlN film [3], or by reactive dc co-sputtering of Al and Cr in a nitrogen gas ambient [4]. The pulsed laser deposition (PLD) technique, due to its capability of reproducing the target stoichiometry in the deposited film, is very suitable for preparation of AlN:Cr films. Prospective AlN:Cr devices and structures formed on Si substrates will be compatible with devices and integrated circuits in contemporary Si planar technology. Because of this it is interesting to prepare AlN:Cr–Si structures and to investigate their properties. For spintronics applications, such as resonant tunneling diodes and
tunneling magnetoresistance structures, the knowledge of charge transport mechanism in AlN:Cr films on Si substrates is of great importance.

In this paper we present results of the study of the charge transport mechanism in the AlN:Cr films prepared on p-Si(100) substrates using the pulsed laser deposition technique. For this purpose, MIS structures with PLD AlN:Cr films were formed and their DC current-voltage (I–V) and admittance (capacitance and conductance) characteristics were measured. Through detailed analysis of these electrical characteristics, the charge transport mechanism was established.

2. Experimental details
Thin AlN films doped with Cr were synthesized on single crystalline Si(100) p-type substrates by pulsed laser deposition [5]. The PLD targets were prepared by homogeneously mixing AlN and Cr (10 at.%) powders followed by pressing and sintering. The films were grown using a KrF* (λ = 248 nm) excimer laser generating pulses of τFWHM ≤ 25 ns at a repetition rate of 10 Hz. During ablation, the target was rotated and translated along two orthogonal axes to avoid punctures and improve the morphology of the deposited films. The incident laser beam was incident at 45°, while ablation plasma was evolving normally to the target surface. The separation distance target–substrate was set to 5 cm. After loading the substrate and target, the deposition chamber was pumped down to a residual pressure of ~10⁻⁴ Pa. Before deposition, the substrate was heated up to 800°C for 1 hour for the purpose of decomposing native SiO₂ on the Si surface. The substrate temperature was kept at 800°C during deposition, providing conditions for crystalline growth of AlN. The laser fluence on the target was 10 J/cm². For the deposition of one film 15 000 subsequent laser pulses were applied yielding a ~300 nm thick films. The AlN:Cr film synthesis was undertaken in vacuum at a residual gas pressure of 10⁻³ Pa.

In order to measure electrical characteristics, MIS capacitors with Al–AlN:Cr–Si structures were formed by vacuum evaporation of Al dots with 0.5 mm diameter on film surface and of a continuous Al film on Si-wafer backside. Room temperature 1 MHz capacitance–voltage (C–V) and conductance–voltage (G–V) measurements were carried out by a digital LCR meter E7-12, while the admittance measurements were carried out by a Tesla BM-507 impedance meter in the frequency range of 500 Hz – 500 kHz.

3. Results and discussion
The DC I–V characteristics of these AlN:Cr MIS structures were measured for electrical fields up to 10⁵ V/cm. The measurement sequence began from 0 V toward negative or positive voltages applied to the contact on the AlN:Cr film surface followed by a voltage reversal toward zero applied bias voltage. The measured I–V curves are given in figure 1. In both forward and reverse bias directions the current is non-ohmic and a hysteresis appears. During the initial voltage passage from zero to minimal negative or maximal positive voltage the current density is larger than that obtained by going back to zero voltage.

![Figure 1. Current density vs. applied voltage characteristics of AlN:Cr MIS structure.](image-url)
For all the four recording directions, when the current density is re-plotted as ln$J$ versus ln$V$ plots the slope of the curves approaches 2, (as an example see figure 2). Such current–voltage dependence is evidence for trap space-charge limited current [6]. In that case, the charge trapped in the insulator is proportional to the applied voltage and the effective velocity of released charge from traps under the applied electric field is also proportional to the voltage. In these circumstances the current is proportional to $V^2$, as we observed.

\[ \Delta V = qN_td/\varepsilon_f, \]

where $\Delta V$ is the voltage difference of up and down curves for a given current density in the $I$–$V$ characteristics (figure 1), $d$ is the film thickness and $\varepsilon_f$ is the film dielectric constant. With $\Delta V = 0.1$ V at $J = -2.43$ A/cm$^2$ and $\varepsilon_f = 31\varepsilon_0$, obtained from the $C$–$V$ measurement, and $d = 300$ nm the density of trapped charge in the AlN:Cr film is estimated as $1.9 \times 10^{17}$ cm$^{-3}$.

![Figure 2](image2.png)

**Figure 2.** ln$J$ vs. ln$V$ plot of current density in reverse down direction on applied voltage of AlN:Cr MIS structure.

The presence of hysteresis in the $I$–$V$ curves is another evidence of trap-connected conductivity in the investigated AlN:Cr films. Starting the $I$–$V$ measurements (so called “up” direction for positive voltages and “down” for negative voltages in figure 1), some of the charges are captured at traps which have time constants in order of tens of minutes, being commensurable to the time for a $I$–$V$ measurement performance. This trapped charge opposes the external electric field of the applied voltage during the voltage passage toward zero. Because of this, in the second voltage passage (so called “down” direction for positive voltages and “up” for negative voltages in figure 1) the current at a given voltage is lower. The density of this trapped charge, $N_t$, can be estimated from the expression $\Delta V = qN_td/\varepsilon_f$, where $\Delta V$ is the voltage difference of up and down curves for a given current density in the $I$–$V$ characteristics (figure 1), $d$ is the film thickness and $\varepsilon_f$ is the film dielectric constant. With $\Delta V = 0.1$ V at $J = -2.43$ A/cm$^2$ and $\varepsilon_f = 31\varepsilon_0$, obtained from the $C$–$V$ measurement, and $d = 300$ nm the density of trapped charge in the AlN:Cr film is estimated as $1.9 \times 10^{17}$ cm$^{-3}$.

![Figure 3](image3.png)

**Figure 3.** $I$–$V$ characteristics with negative differential resistance of AlN:Cr MIS structure.
In figure 3 the change of the current density in the forward direction with the voltage applied to the AlN:Cr MIS structure is shown. These dependences were obtained by registering the current alteration through the structure during 15–20 minutes, as the total voltage applied to this structure and the current measuring serial resistor was kept constant. The current density decreases when the electric field increases. This decrease is less than 1% at electric field around $1.6 \times 10^3$ V/cm and it is approximately 10% at electric field around $10^5$ V/cm. In [7] for electric fields above $2 \times 10^6$ V/cm the negative differential mobility in AlN films has been calculated and it has been attributed to polar optical phonon scattering of electrons at these high electric fields. However, in our AlN:Cr–Si structures the negative differential resistance, observed at electric fields ranging from $1.6 \times 10^3$ to $10^5$ V/cm differs from that discussed in [7]. The presence of traps in the AlN:Cr films with very long capture and emission time-constants, in the order of minutes, can be the reason for the observed negative differential resistance. After applying the voltage to the AlN:Cr–Si structure, additional electrons are captured at these traps and this additional charge opposes the external electric field leading to the observed negative differential resistance. Decrease of the electric field $\Delta E$ can be expressed as $\Delta E = qN_{t,a}d/\varepsilon_f$, where $N_{t,a}$ is the density of additional charges at traps with long time constants. Therefore, at 15% decrease of $10^5$ V/cm electric field, with $\varepsilon_f = 31 \varepsilon_0$ and $d = 300$ nm the estimated $N_{t,a}$ density is $8.6 \times 10^{15}$ cm$^{-3}$. Both densities of trapped charges $N_t$ and $N_{t,a}$ are reasonable. The difference between these values arise from the fact that different traps are responsible for the hysteresis in the $I$–$V$ characteristics and for the negative differential resistance of the studied AlN:Cr–Si structures.

The impedance of the AlN:Cr MIS structure was measured at test signal voltages in the 500 Hz – 500 kHz frequency range without applying bias voltage. The values of $Re Z = |Z|\cos \phi$ and $Im Z = |Z|\sin \phi$ are plotted in figure 4. As it is seen, the maximal values of $Re Z = 124$–$128$ Ohm correspond to values of $Im Z = 115$–$152$ Ohm, measured at 1 and 2 kHz. From this follows that $\omega \tau = 1$, where $\tau$ is the response time of traps related with impedance data and, it is equal to $\tau = 80 \mu$s at 2 kHz.

The $C$–$V$ and $G$–$V$ characteristics of the AlN:Cr–Si MIS structures, measured at 1 MHz are presented in figures 5 and 6, respectively. The sweep direction is given with an arrow, starting from negative to positive voltages (“up”) and contrariwise (“down”). One may notice that up and down $C$–$V$ curves in the sequence from $–20$ V to $20$ V and the reversal to $–20$ V almost coincide with each other. At applied voltages below $–7.2$ V and above $7$ V both kinds of characteristics saturate, which one may expect for a MIS structure. The observed saturation in accumulation and deep inversion regions is due to the complete filling of traps in the AlN:Cr film at these bias voltages.
In the voltage region between \(-7.2\) V and 1.3 V a bump appears in the C–V curves. Similar capacitance feature is observed in many semiconductor structures and it is connected with the back-to-back Schottky model of depletion regions on both sides of the grain boundary [8]. But the sharp transition to the accumulation capacitance at \(-7.2\) V together with the sharp decrease of the capacitance above 1.3 V indicate that this capacitance bump is connected with traps, which are able to respond to the bias voltage at 1 MHz. From the shape of the G–V curves (figure 6) one can conclude that the traps, responding to 1 MHz test voltages, are filled with charge carriers to different extent in the voltage region between \(-7.2\) V and 7 V and, they become completely filled in the regions, where the capacitance and the conductance are constant. There are not empty traps below the charge carrier quasi-Fermi levels in these AlN:Cr films.

The permittivity \(\varepsilon_f\) of the AlN:Cr film was calculated from the capacitance value in accumulation and it was equal to \(31\varepsilon_0\). This value is higher than the value \(\varepsilon_f \approx 10\varepsilon_0\), estimated from the optical measurements of AlN films in [9]. In [10] \(\varepsilon_f\) has reached even \(400\varepsilon_0\) and this high value has been explained by defects related to nitrogen vacancies in AlN films. Therefore, it is reasonable that due to the high density of traps in the AlN:Cr films the trapped charges contribute to additional capacitance in accumulation regime.

4. Conclusions
It has been established that the current through the AlN:Cr–Si MIS structures in both forward and reverse directions is proportional to square of applied voltage and it is governed by the space charge at filled traps in the AlN:Cr films. The density of these traps with long emission time-constants is estimated from the hysteresis of the \(I–V\) curves and it is \(1.9 \times 10^{17}\) cm\(^{-3}\). The density of traps with long capture time-constants, which are responsible for the observed negative differential resistance, is \(8.6 \times 10^{15}\) cm\(^{-3}\). Both capacitance and conductance values saturate in accumulation and deep inversion regime revealing that the traps are completely filled below the quasi-Fermi level in these AlN:Cr films.

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