Leakage current conduction of pulsed excimer laser ablated $BaBi_2Nb_2O_9$ thin films

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The leakage current behavior of the $BaBi_2Nb_2O_9$ (BBN) thin films was investigated over a wide range of temperatures. The current density, calculated from current–voltage ($I-V$) characteristics at room temperature, was 4.02×10^{-9} A/cm^2 at an electric field of 3×10^5 V/m. The $I-V$ characteristics of the films showed ohmic behavior for electric field strength lower than 1 MV/m. Nonlinearity in the current density–voltage ($J-V$) behavior was observed at an electric field above 1 MV/m. Different conduction mechanisms were brought into picture to explain the $I-V$ characteristics of BBN thin films. The $J-V$ behavior of BBN thin films was found to follow the Lampert's theory of space charge limited conduction in an insulator with traps. Three different regions, i.e., ohmic, trap filled limited, Child's law were explicitly observed in $J-V$ characteristics. The activation energies in the ohmic region calculated from the Arrhenius plot were 0.46, 0.48, and 0.51 eV, respectively. These energies were attributed to the shallow traps, distributed near the conduction band edge in the forbidden gap of the materials.

I. INTRODUCTION

Bi-layered structure compounds have been found to be the most suitable alternative material for $Pb(ZrTi)O_3$ (PZT) because of their fatigue free behavior in data storage devices, e.g., nonvolatile random access memories, smart card, etc. The performance of the data storage devices depends on their ability to retain the stored charge of the active materials, which store the information in terms of charge.[1] Various issues such as polarization switching, fatigue and imprint have been extensively investigated for many ferroelectric materials before integrating them onto the integrated circuit. Another significant feature, which is of crucial importance for the quality and reliability of the ferroelectric devices, is low leakage current. Particularly for thin films, where a nominal voltage across the sample produces a very high electric field, the mechanism of charge transport becomes very complicated. In addition to electrical measurements all the dielectric measurements are concerned with the movement of the charge, dc conduction (σ_0) must appear in the dielectric results because the measuring instrument cannot discriminate between true dielectric response and that combined with dc contribution. The study of the conduction mechanism may help in resolving these components. Several types of conduction mechanism including Poole–Frenkel emission, space charge limited current, and Schottky barrier limited conduction mostly in PZT thin films have been addressed in the literature.[2–7] But in the case of the Bi-layered structure compounds, which are also considered as a potential candidate for data storage devices, the leakage current behavior has not been well studied. Our intention is to elucidate the true leakage current behavior of pulsed laser ablated $BaBi_2Nb_2O_9$ (BBN) thin films and to find a particular mechanism suitable for explaining the present data.

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II. EXPERIMENT

The BBN thin films were deposited by a Lambda Physik (KrF) excimer laser ($\lambda = 248$ nm) on a platinum coated silicon substrate. The frequency and the energy fluence of the laser beam during deposition was kept at 5 Hz and 4 J/cm^2, respectively. The distance between the target and the substrate was maintained at 3 cm. Prior to the deposition process the base pressure was brought down to 1.0×10^{-5} Torr. The substrate temperature during deposition was maintained at 400 °C. A relatively high oxygen pressure (100–250 mTorr) was used to maintain compatibility between the number of oxygen molecules impinging on the substrate surface per unit time in this range of pressure and ablated ions/atoms arriving from the target per second. The as-deposited films were annealed at different temperatures (650–750 °C) for 30 min in a quartz tube furnace under flowing oxygen. The crystallographic structure of the film and the target were confirmed by x-ray diffraction using Cu K_α radiation. A semiquantitative analysis of the film composition was performed by energy-dispersive analysis of x rays. Gold top electrodes of 1000 Å thickness were deposited at room temperature by a thermal evaporation process through a shadow mask of area 1.96×10^{-3} cm^2. The thickness of the film was confirmed using an optical spectrometer (Filmetrics F-20 thin-film measurement system). The thickness of films varied from 0.2 to 0.9 μm. Details of structural and dielectric properties of BBN thin films are available elsewhere.[8,9] The leakage current characteristics of BBN thin films were studied using a Keithley 236 source measure unit at various temperatures after stabilizing the temperature for 10 min prior to

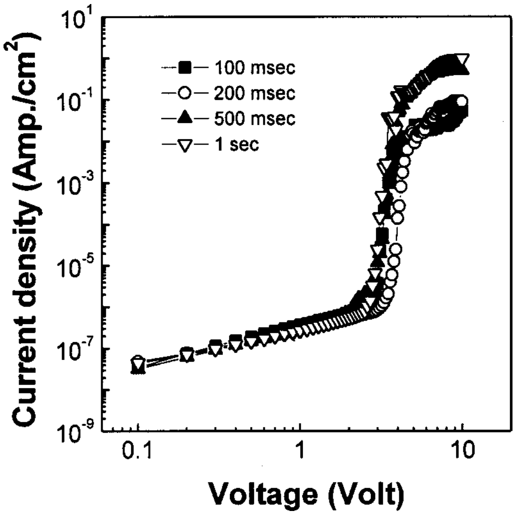

FIG. 1. Room temperature $I-V$ characteristics of BBN thin films with different delay times.

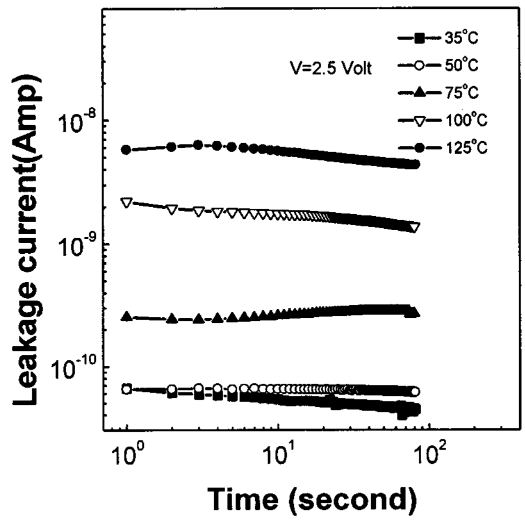

FIG. 2. Current–time $(I-t)$ response at different temperature with a constant bias.

each measurement. All the measurements were taken under a step voltage of 0.1 V and delay time of 100 ms.

III. RESULTS AND DISCUSSION

When a voltage is applied to the ferroelectric capacitor the current flowing through the external circuits comprised of two components: polarization contribution and leakage contribution. The polarization current shows a transient behavior until a certain time and might contribute to the overall response of current–voltage $(I-V)$ behavior if the measurements are taken within the transient time. To perceive any interference of the transient response into the measured value we performed the $I-V$ measurement at different delay times, as shown in Fig. 1. The results did not show any significant change, confirming that the experimentally measured current for 100 ms delay times was the true leakage behavior of the sample. This was once again confirmed from the $I-t$ behavior (Fig. 2) at constant voltage and different temperatures. The $I-t$ response was started from 0.1 s because of experimental limitation. The nature of the curve clearly indicates that the time domain, which was used for $I-V$ measurements, was beyond the transient regime of our sample. Therefore we can conclude that the $I-V$ response was the true leakage behavior of the BBN sample. Dietz and Waser did a detailed analysis of transient response and leakage current of dielectric thin films.[10] The leakage current behavior of the BBN thin films is shown in Fig. 3. At the low field region the current density–voltage $(J-V)$ characteristics are ohmic in nature. Different conduction mechanisms, e.g., Schottky,[11] Poole–Frenkel,[12] and space charge limited conduction[13] were verified to explain the true nature of charge transport phenomena in BBN thin films.

Schottky emission mechanism is an electrode limited conduction, where the entire leakage current is dominated by the Schottky barrier generated at the interface of the electrode and film. The electrode-limited current density (so called Schottky current) upon application of the electric field behaves according to the following equation:

$$J=AT^2 \exp\left[\frac{(\Phi_0-\beta_s E^{1/2})}{kT}\right], \tag{1}$$

where $A=(4\pi e m^* k^2/h^3)$ is the Richardson constant, and Φ_0 is the barrier height generated due to the work function difference between metal and insulator. $\beta_s=(e^3/4\pi\varepsilon_0\varepsilon_r)^{1/2}$, where ε_r is the high frequency dielectric constant, ε_0 is the free space permittivity, k is the Boltzmann constant, h is the Plank constant, m^* is the effective mass of the electron, and E is the electric field across the film.

To verify the Schottky dominated conduction in BBN thin films, we plotted J/T^2 versus $E^{1/2}$ in the semilog scale, which was supposed to be a straight line. The Schottky plot for BBN thin films is shown in Fig. 4(a), where nonlinearity was observed at the high field (>40 kV/cm) region. So, concentrating only in the lower field region, where the curves seemed to be a straight line, the high frequency dielectric

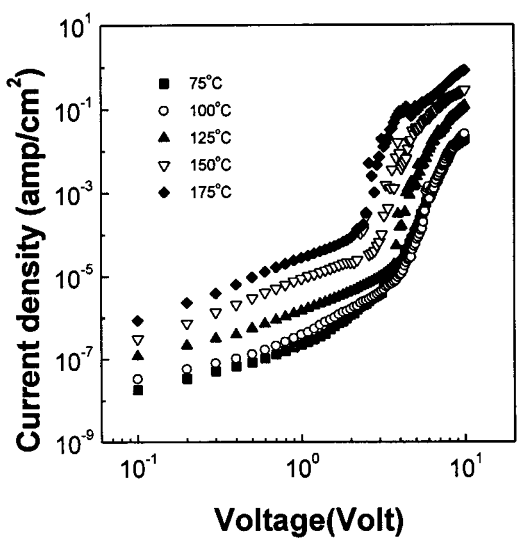

FIG. 3. $I-V$ characteristics of BBN thin films at different temperatures.

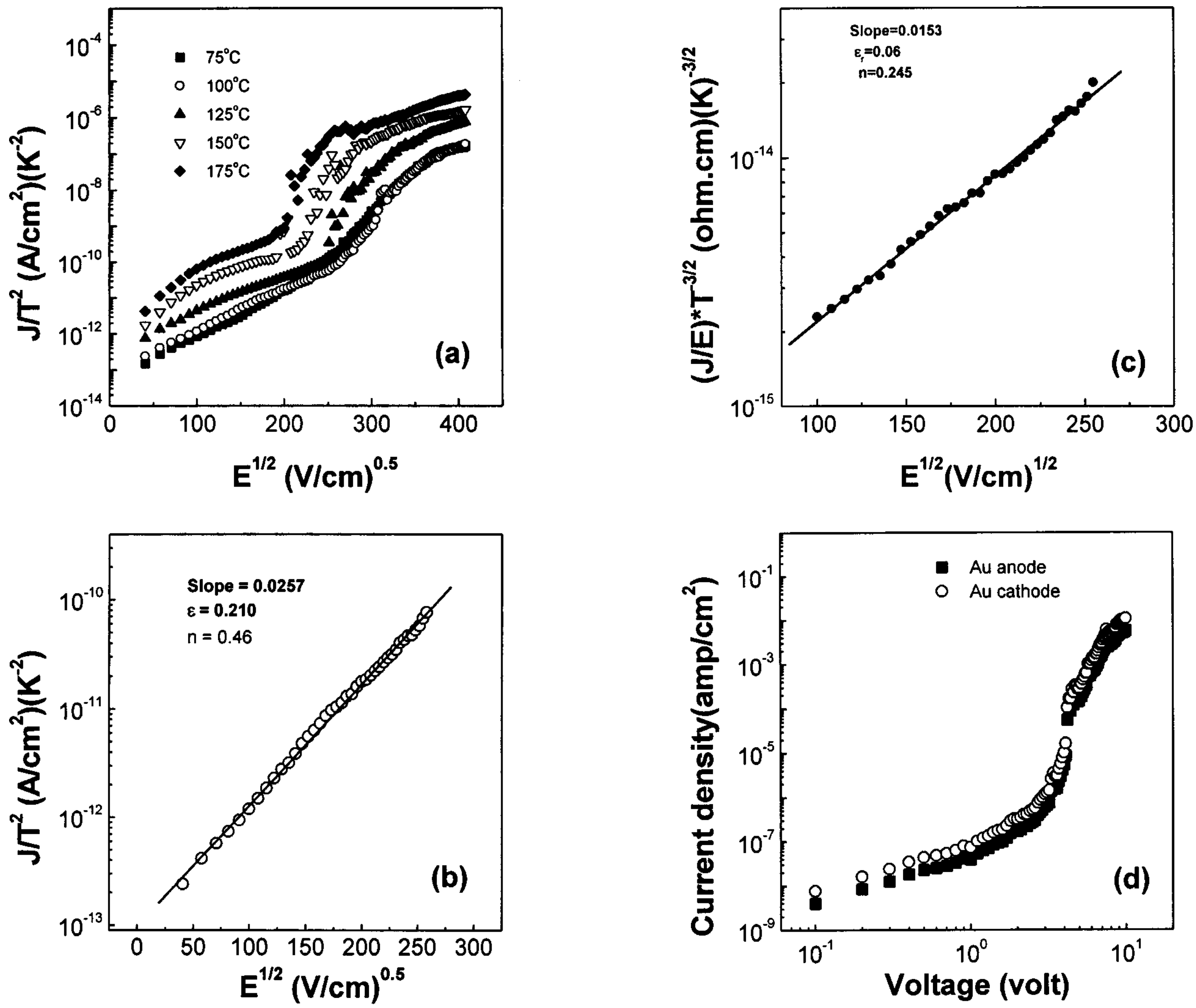

FIG. 4. (a) Schottky plot for BBN thin films at various temperature. (b) J/T^2 vs $E^{1/2}$ plot of BBN thin films at 100 °C. (c) Modified Schottky plot at 100 °C. (d) Influence of electrode polarity on $J-V$ response.

constant was calculated from the slope of $\ln(J/T^2)$ versus $E^{1/2}$ plot at 100 °C [shown in Fig. 4(b)]. The calculated dielectric constant and refractive index ($\varepsilon_r = n^2$) are 0.21 and 0.46, respectively. These values are 1 order less than the actual values. To justify the validity of the Schottky mechanism the modified equation, as proposed by Sufi Zafar,[14] was also considered. The modified Schottky equation is given by

$$J = BT^{3/2}E \exp\left[\frac{(\Phi_0 - \beta_s E^{1/2})}{kT}\right], \tag{2}$$

where B is a constant dependent on the mobility and effective mass of the electron. Figure 4(c) shows the modified Schottky plot at 100 °C and the value of ε_r (=0.06) and n (=0.245) were calculated from the slope. These values also did not seem to agree with the actual values. A third verification of the electrode influence on the charge injection was done by changing the polarity of the electrodes. Figure 4(d) shows the room temperature leakage current behavior of BBN thin films with Au as a cathode and anode, respectively. No significant variation of leakage current was observed in two different cases. These inconsistencies in the present results led us to conclude that the Schottky mechanism may not be useful in explaining the charge transport behavior of BBN thin films. However, the best method to investigate the validity of the Schottky emission is to change the electrode materials, which was reported by Dietz *et al.*[15] and Dey *et al.*[16] The influence of the different top electrodes on the leakage current behavior of BBN thin films is in progress and will be reported on later.

Contrary to the electrode-limited conduction, the Poole–Frenkel mechanism is a bulk limited charge transport in the insulating thin film.[17,18] It is recognized in terms of lowering the potential barrier of the trap levels in the application of an external electric field. These trap levels are assumed to be positively charged when they are empty. Hence, due to Coulomb force the charge carriers (thermally generated and injected) are captured by the trap levels and consequently these trap levels become neutral. Functional dependence of the Poole–Frenkel (PF) dominated conduction is given by the following equation:

$$J = CE \exp\left[\frac{(\Phi_0 - \beta_f E^{1/2})}{kT}\right], \tag{3}$$

where $\beta_f = (e^3/\pi\varepsilon_0\varepsilon_r)^{1/2}$, and C is a constant in terms of the

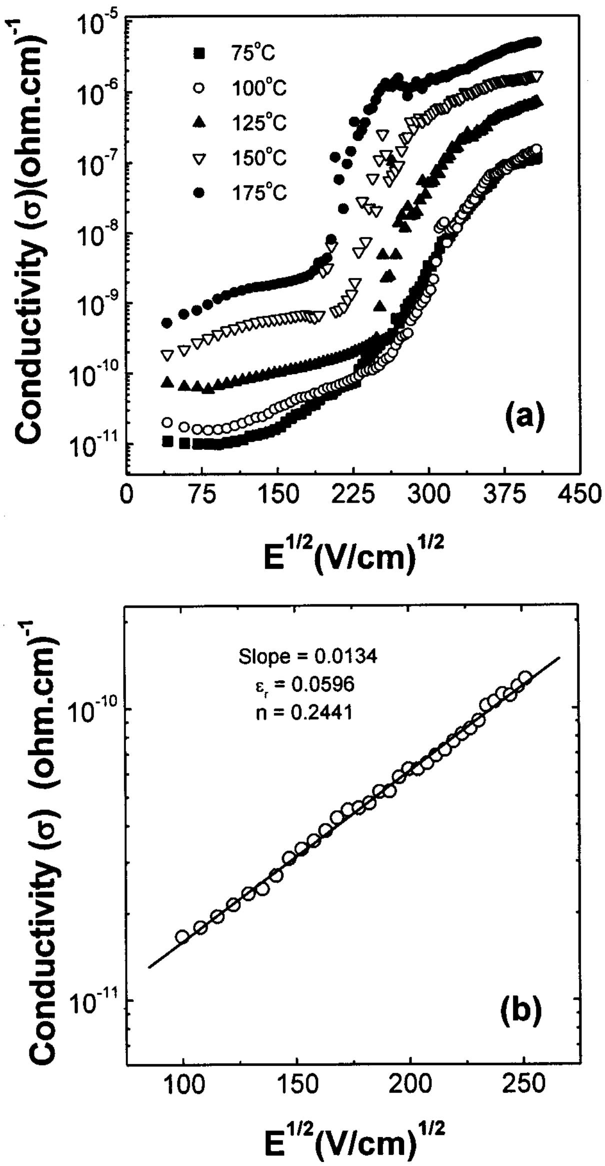

FIG. 5. (a) Poole–Frenkel plot for BBN thin films at various temperature. (b) J/E vs $E^{1/2}$ plot of BBN thin films at 100 °C.

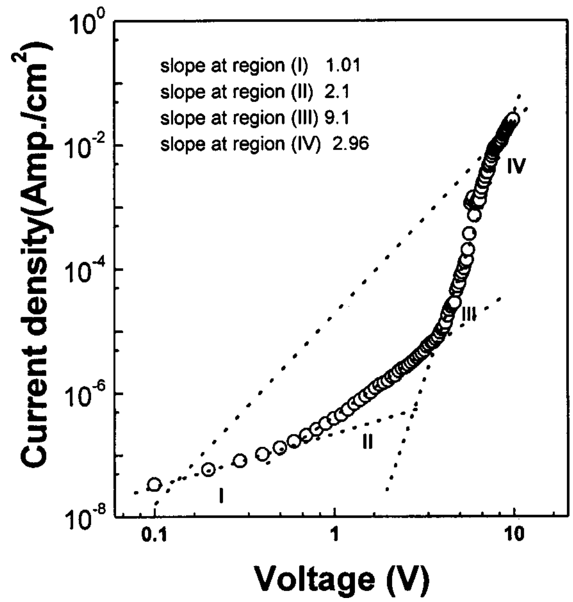

FIG. 6. $J-V$ plot for BBN thin film at 100 °C.

trapping density in the insulator. Therefore, identifying the PF conduction in the thin films is the same as the Schottky type. Figure 5(a) shows the conductivity ($\sigma = J/E$) versus $E^{1/2}$ plot for BBN thin films. High frequency dielectric constant ε_r calculated from the slope β_f [shown in Fig. 5(b)] was found to be 0.0596, while the corresponding refractive index was 0.244. These results are again contrary to the actual values.

Finally, a space charge limited (SCL) conduction mechanism was found to be appropriate to explain the charge transport behavior in BBN thin films. The nature of the curve indicates that the dc conduction in BBN thin films follow the Lampert's theory of SCL-current. The three interesting regions, which are governed by three mechanisms, are clearly distinguishable in Fig. 6. The $J-V$ characteristics are confined with in a triangle in the $\log J$–$\log V$ plane bounded by the three limited curves: Ohm's law, trap filled limit and Child's law ($J \alpha V^2$). A threshold voltage is required to get the trap filled limited (TFL) region, which shows the sudden increase in current with a very high rate. So, the overall view of the leakage current can be described as[19–21]

$$J = J_{\text{Ohm}} + J_{\text{TFL}} + J_{\text{Child}}, \tag{4}$$

with

$$J_{\text{Ohm}} = en\mu\left(\frac{V}{d}\right), \quad J_{\text{TFL}} = K\left(\frac{V^{(l+1)}}{d^{(2l+1)}}\right),$$

and

$$J_{\text{Child}} = 9\varepsilon\mu\left(\frac{V^2}{8d^3}\right),$$

where e is the electronic charge, μ is the mobility of the of the charge, n is the concentration of the free charge carrier in the insulator, V is the voltage applied across films of thickness d, $l = (T_t/T)$, T_t is the temperature dependent parameters characterizing trap distribution, and T is the absolute temperature.

At low applied voltages, $J-V$ characteristics followed Ohm's law because the density of the thermally generated carriers in the films was predominant over the injected charge carrier. The activation energies calculated from the ohmic region of the $J-V$ characteristics were 0.46, 0.48, and 0.51 eV, respectively, shown in Fig. 7. These activation energies were attributed to the shallow traps, existing near the conduction band of the BBN thin films. The variation of the activation energy implies that the traps are distributed in the forbidden region, but the type of distribution is not yet clear. The onset of the departure from Ohm's law or the onset of the SCL conduction takes place at the applied voltage reaches

$$V_{\text{on}} = \frac{8end^2}{9\varepsilon\theta_a}, \tag{5}$$

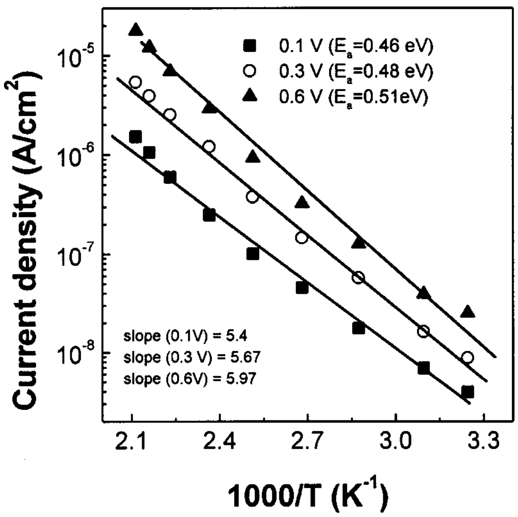

FIG. 7. Arrhenius plots of BBN thin films calculated from ohmic region of $J-V$ characteristics.

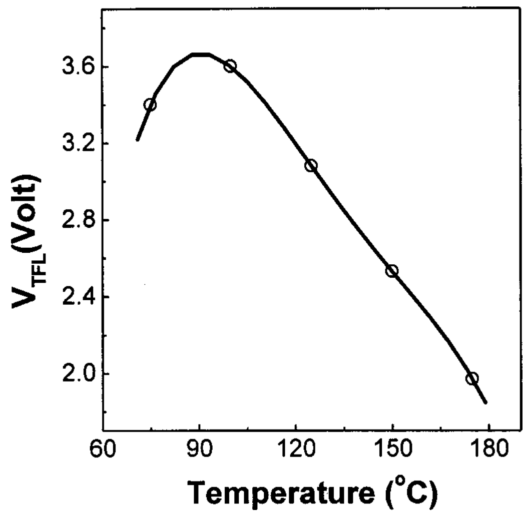

FIG. 9. Variation of V_{TFL} with temperature

where ε is the dielectric constant of the capacitor, $\theta_a = (N_c/g_n H_d)\exp(-E_t/kT)$, the ratio of the free carrier density to total carrier (free and trapped), H_d is the density traps, N_c is the density of the states in the conduction band, g_n is the degeneracy of the energy state in the conduction band, and E_t is the energy level below the minimum of the conduction edge. The variation of the V_{on} with temperature for BBN thin films is shown in Fig. 8. The figure illustrates that the onset voltage V_{on} of SCL conduction increases with temperature, which is obvious because the density of the thermally generated carrier increases with temperature. At the region of transition from the ohmic to SCL, the carrier transit time $\tau_c = d^2/\mu V_{\mathrm{on}}$ becomes equal to the Ohmic relaxation time $\tau_d = \varepsilon/en\mu$,[19] where μ is the mobility of the charge carriers. The sudden jump in the $J-V$ characteristics at a particular voltage implies the crossover from ohmic to TFL conduction. At this voltage the injected carriers saturate all the trap levels. After the saturation of defect levels additional excess charges appear in the conduction band, which is responsible for a sudden increase in current. Figure 9 shows the variation of onset voltage of the trapped filled limited region V_{TFL} with temperature. Since the thermal generation of the charge carrier increases with temperature, relatively lower voltage is required to fill all the trap levels at higher temperature. For the voltage $V > V_{\mathrm{TFL}}$ the current is fully controlled by the space charge, which limits the further injection of free carriers in the sample. Square law dependence of the current ($J \sim V^2$, Child's law) in the voltage range $V > 2V_{\mathrm{TFL}}$ is the consequence of the space charge controlled current. Figure 10 shows the experimental dependence of leakage current with the film thickness, which is in agreement with the theoretical prediction $I \sim d^{-3}$, where d is the thickness of the film. Such agreement of the experimental results with the predicted theory ascertains the validity of SCL conduction to the leakage current behavior of present BBN thin films.

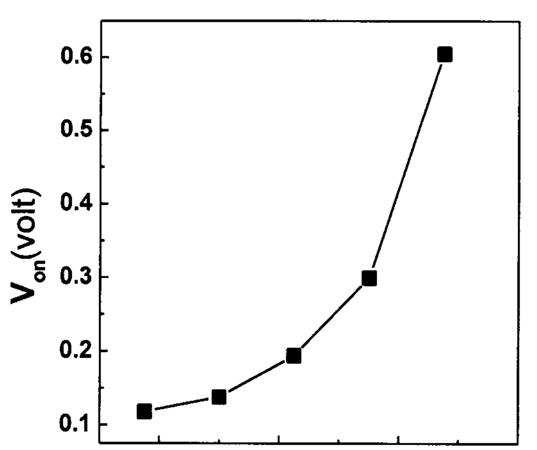

FIG. 8. Temperature dependence of onset voltage V_{on} to the space charge region.

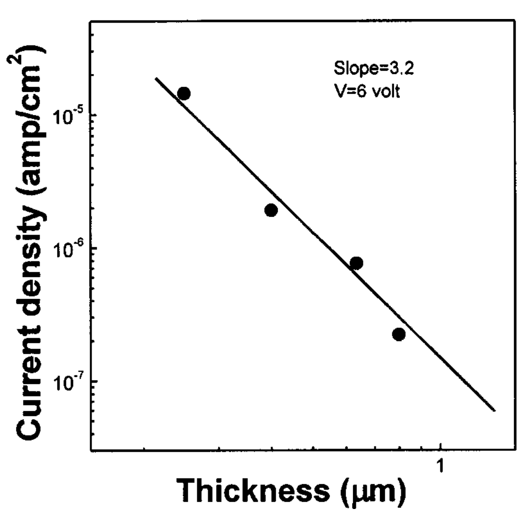

FIG. 10. Thickness dependence of leakage current of BBN thin films at room temperature.

IV. CONCLUSIONS

We investigated the leakage current behavior of the BBN thin films at various temperatures. The $I-V$ characteristics followed Lampert's theory of SCL current. Three regions: Ohmic, trapped filled limited, and Child's law were clearly observed in the $J-V$ characteristics. Only the existence of shallow traps was found to be present in the sample within the experimental conditions. The trap energies calculated from the ohmic region of $J-V$ characteristics were 0.46, 0.48, and 0.51 eV, respectively. The onset of the SCL region was found to increase with temperature, whereas V_{TFL} decreased with temperature.

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