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Joule heating and breakdown effects in KNO_3 thin layers

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The current–voltage (I–V) characteristics of KNO_3 single-crystal thin layers of thickness range 35 to 145 μm have been studied and the critical voltage V_c and breakdown voltage V_b determined. The results are interpreted taking into account Joule heating. Experimental evidence for the existence of Joule heating in KNO_3 thin layers has been found. A semi-theoretical approach to the data involving a law of cooling has enabled the activation energy associated with the current increase to be estimated and found to be $\phi = 0.05$ eV. The effect of externally heating the layer indicates a change in the layer current at $T_c = 130°$ C. This suggests phase changes in KNO_3 thin layers, and this is strongly supported by differential thermal analysis. It is therefore concluded that the I–V characteristics of KNO_3 thin layers are strongly influenced by Joule heating which causes changes in the KNO_3 phase. A discussion is given on which type and mechanism of electrical breakdown is present in thin layers.

1. Introduction

In recent years, much attention has been centred on studies of the structure and properties of single-crystal thin layers [1, 2]. Such studies are of interest both from the scientific and technological standpoints. It is quite obvious that a proper interpretation of the physical properties of thin layers is dependent not only on adequate information regarding the arrangement of atoms in the layers but also on knowledge regarding the lattice defects existing in them. Sometimes thin insulating layers are used as surface passivation insulation between conductors and as dielectrics in capacitors. In all these applications it is mandatory that the insulating layer retains its insulating properties with time and stress. Consequently, the electrical breakdown strength has received great attention. In this paper we report the results of the current–voltage (I–V) characteristics of KNO_3 thin-layer crystals, taking into account the influence of Joule heating. Also, the scope of this work is to further clarify some aspects of the breakdown phenomena in thin insulating layers. An approach like this has been very rewarding in the study of high-voltage insulating systems. It is to be hoped that this approach will be equally useful in microelectronic devices.

There are three well-known forms of solid potassium nitrate at atmospheric pressure [3]. Phase I is stable between 128° C and the melting point, 334° C, whereas Phase II is stable below 128° C [3–5]. On cooling, Phase I transforms to the metastable ferroelectric Phase III at 123° C under certain conditions [6, 7]. Phase III is stable at higher pressures at which other modifications are also formed [8]. At ordinary pressures Phase III has been found to transform apparently irreversibly into Phase II, starting at about 115° C [4, 5].

Phase II has the orthorhombic aragonite structure [9, 10]. The high-temperature Phase I has been reported to have a rhombohedral structure with the nitrate groups orientationally disordered in calcite-type positions. Phase III is reported to have an ordered rhombohedral structure with the nitrate groups in aragonite-type positions [11]. The structures of the three phases and their space groups are shown in Fig. 1. The structural properties of KNO_3 have been reviewed and discussed in relation to thermodynamic quantities [12].

2. Experimental procedure

A thin layer of KNO_3 single crystal was prepared by the method explained in detail in previously published work [1, 13]. A thin layer cross-section looks like the sketch shown in Fig. 2. A metallic common bottom electrode of tin oxide is deposited on one side of an insulating glass plate. On top of this a thin layer of KNO_3 single crystal, usually 35 to 145 μm thick, is deposited. This layer forms the dielectric single-crystal section to be tested. A set of metallic spots 5 mm in

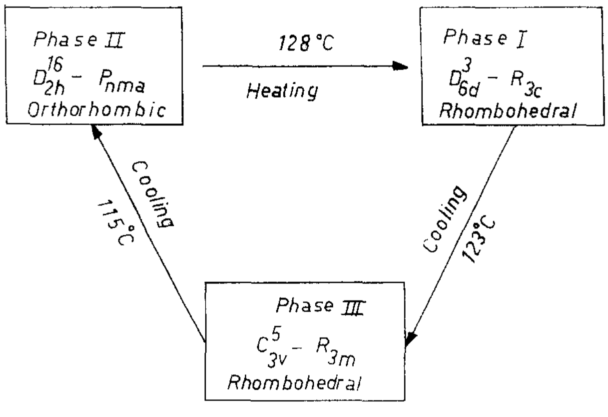

Figure 1 Phase relations in KNO_3.

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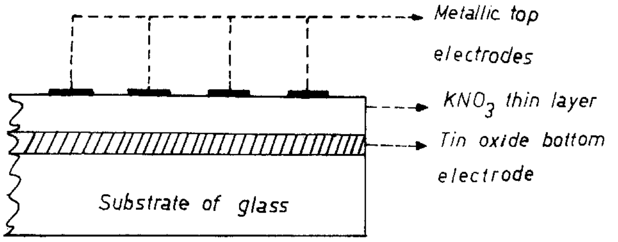

Figure 2 Cross-section of KNO_3 thin-layer single crystal prepared on a glass substrate.

diameter acts as the top electrodes of the layer. All of these spots are silver thin films and can be prepared by vacuum evaporation or sputtering.

3. Results and discussion

Fig. 3 shows a graph of the d.c. I–V characteristic of a KNO_3 thin layer 35 μm in thickness in which two straight lines of different slopes could be observed. The slope difference starts at a critical voltage V_c (5 V for a layer of 35 μm thickness). Also as the applied voltage increases, one can reach a breakdown voltage V_b at which a sudden sharp rise in electric current takes place. Thus, as the applied voltage across the layer is increased beyond a certain level, the dielectric layer is locally destroyed and the capacitor is, at least temporarily, short-circuited. This phenomenon is well known and widely discussed in general dielectrics [14, 15]. Several models for such physical phenomena in solids have been provided [16–18]. The currents at V_c and V_b are controlled by two different mechanisms. This will be explained and discussed later. Fig. 4 shows the I–V characteristics of KNO_3 single-crystal thin layers at different layer thicknesses. As is clear, for each layer thickness there is a certain critical voltage V_c and breakdown voltage V_b. The variation of V_b and V_c with layer thickness is shown in Fig. 5.

3.1. The dependence of d.c. current on Joule heating

Graphs of I against V for d.c. data gave straight lines up to a certain voltage after the usual initial ohmic range (Fig. 6). However, above this critical voltage V_c, the data lay on a straight line of increased slope. This suggested the possibility of Joule heating causing a current rise when the applied voltage exceeded a critical value V_c.

In order to interpret the results it was necessary to make certain assumptions, which led to an equation which could then be tested using the experimental data.

As shown in Fig. 6, the data at voltages less than V_c fit a square law. At a voltage greater than V_c, it is supposed that the current is raised to a value I_2 corresponding to a layer temperature T_2, whereas under the condition $V < V_c$ the steady current has a value I_1, at a layer temperature T_1 (in general $T_1 \neq T_0$, where T_0 is the room temperature). Hence

$$I_2 = aV^2 \exp\left(\frac{-\phi}{2kT_2}\right)$$

and

$$I_1 = aV^2 \exp\left(\frac{-\phi}{2kT_1}\right)$$

where a is a constant.

It is supposed here that there is an exponential dependence on temperature with activation energy ϕ; then

$$\ln\left(\frac{I_2}{I_1}\right) = -\frac{\phi}{2k}\left(\frac{1}{T_1} - \frac{1}{T_2}\right) \qquad (1)$$

assuming a law of cooling of the simple form

$$I_2V_2 = M(T_2 - T_0) \qquad (2)$$

where M is a constant and I_2V_2 is the energy per second. The above equation is Newton's law of cooling and supposes that heat is lost steadily to the surroundings by conduction. A similar approach has been used [19] in discussing thermal breakdown in insulators. Substituting for T_2 from Equation 2 into Equation 1,

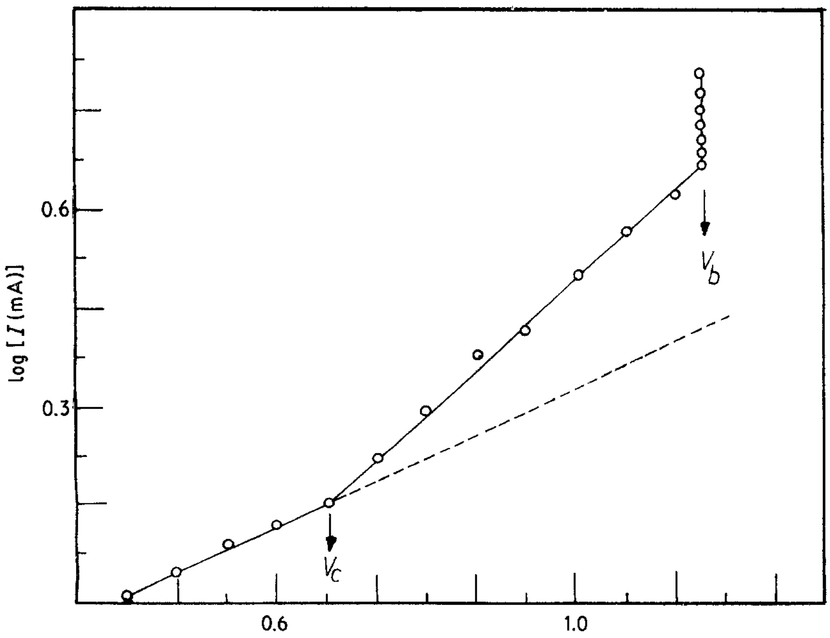

Figure 3 Graph of log I–log V characteristic of thin KNO_3 layer (d = 35 μm).

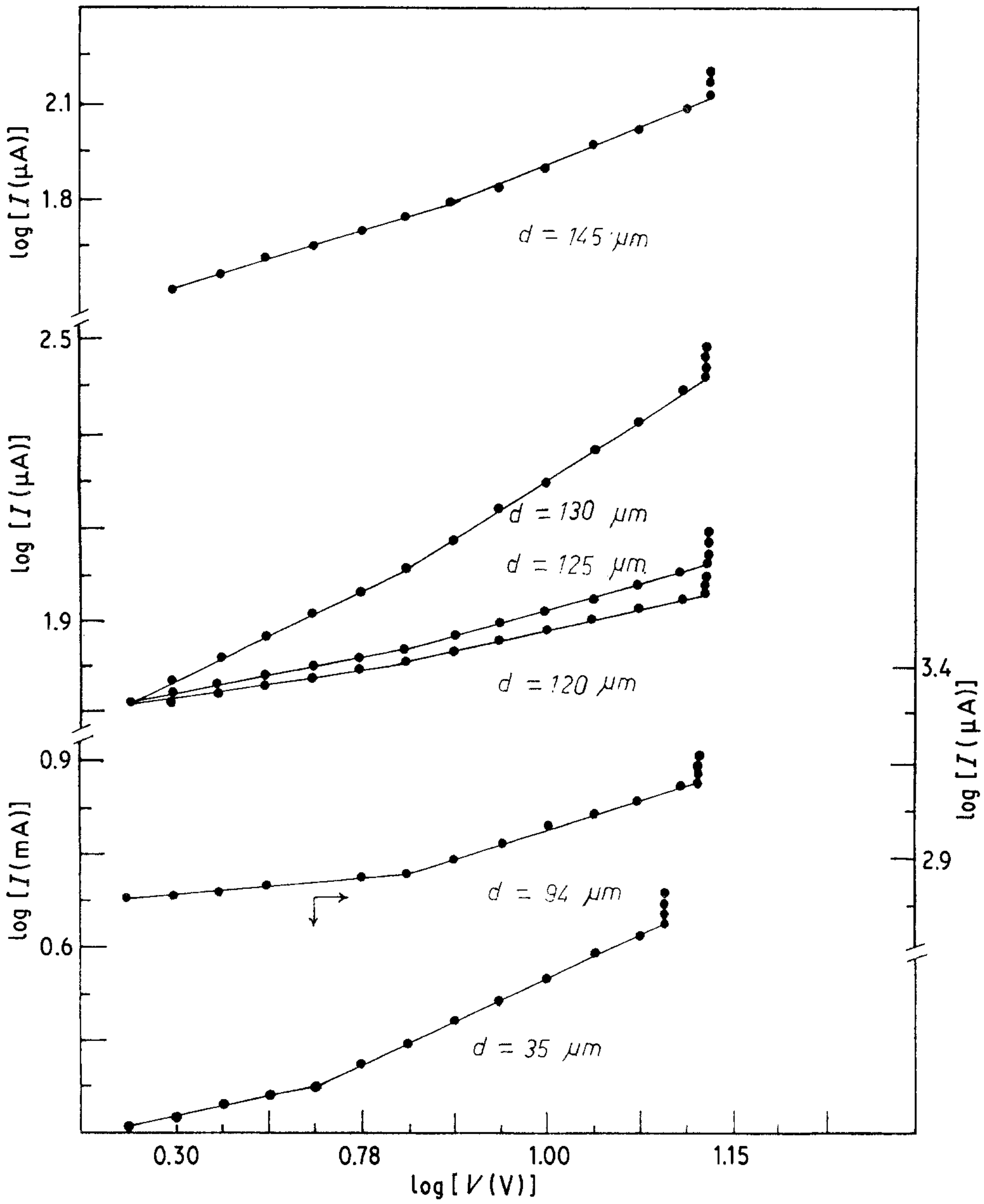

Figure 4 Graph of log I–log V for KNO$_3$ thin layers of various thicknesses.

we have

$$\frac{1}{\ln(I_2/I_1)} = \frac{2kT_1}{\phi}\left(\frac{I_2V + MT_0}{I_2V + M(T_0 - T_1)}\right) \quad (3)$$

When $T_1 = T_0$, then Equation 3 becomes

$$\frac{1}{\ln(I_2/I_1)} = \frac{2kT_0}{\phi} + \frac{2kT_0^2M}{\phi}\left(\frac{1}{I_2V}\right)$$

The above equation represents a straight line. It seems more convenient to plot a slightly different form of the equation, namely

$$\frac{I_2V}{\ln(I_2/I_1)} = \frac{2kT_0}{\phi}I_2V + \frac{2kT_0^2M}{\phi} \quad (4)$$

Inserting the data into Equation 4, a satisfactory straight line results (Fig. 7). The slope enables the activation energy to be calculated as $\phi = 0.05\,\text{eV}$. Hence M may be deduced. To estimate the temperature T_c at the critical voltage V_c, consider $(I_2V)_c$ as the

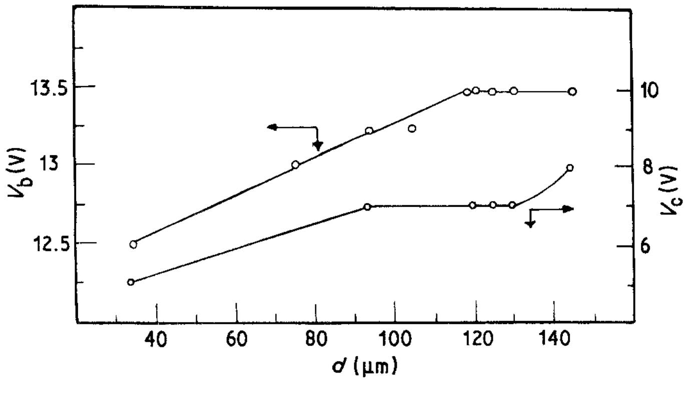

Figure 5 Variation of breakdown voltage V_b and critical voltage V_c with the layer thickness.

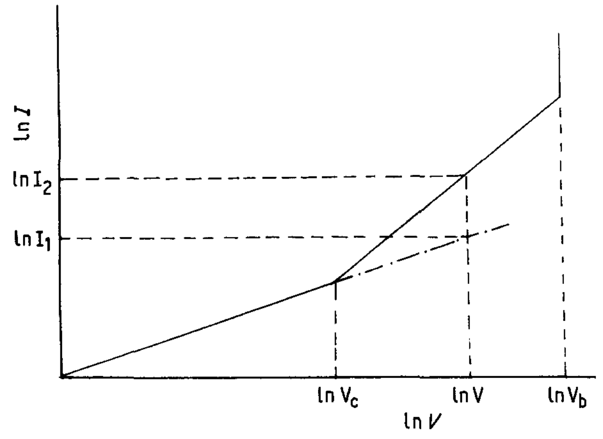

Figure 6 Schematic form of the log I–log V characteristic.

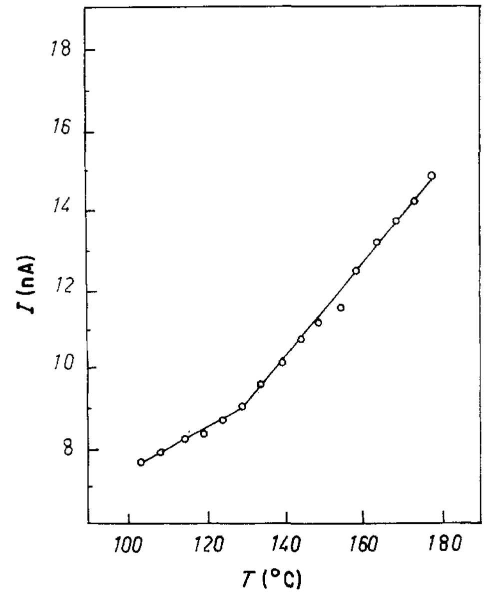

Figure 8 Variation of the electric current with temperature for KNO_3 thin layer.

corresponding critical power; then from Equation 2

$$(I_2V)_c = M(T_c - T_0)$$

Hence $T_c = 130°C$.

To gain some insight into the significance of T_c, measurements of d.c. current at a fixed voltage as a function of temperature were made. The results are shown in Fig. 8, and indicate a change in layer conductance as a function of temperature at ~ 130° C in the heating cycle. One can suggest that T_c may be identified as a phase transition temperature. An excellent confirmation of this suggestion is obtained by differential thermal analysis (DTA) curves carried out for KNO_3 thin layers [20] (Fig. 9), in which a clear phase transformation is recorded at 130° C. Preliminary examination of the I–V data reported by other workers suggests that Joule heating is a widespread feature of direct-current electroluminescence I–V data [21, 22].

The value of the critical power $(I_2V)_c$ varies considerably from sample to sample. This is related to the variation in M from sample to sample. If a test area lies close to the substrate edge, its ability to cool is expected to be larger than if it is more centrally placed. Similarly, if the sample and the substrate size are relatively small $(I_2V)_c$ would be expected to be small.

It seems likely from the evidence for Joule heating that changes in electrical properties of the KNO_3 thin layers phase occur as the applied voltage is raised. It is therefore concluded that the I–V characteristics of KNO_3 are strongly influenced by Joule heating which causes changes in the thin-layer phase.

3.2. Breakdown characteristics

Fig. 10 shows the detailed steps of breakdown for a KNO_3 single-crystal thin layer. Notice that as the voltage is increased beyond V_c, the current remain extremely small up to the knee of the curve (Fig. 10). At this point, the breakdown effect begins. The layer resistance begins to decrease as the layer current increases rapidly. From the bottom of the knee, the breakdown voltage V_b remains essentially constant. The layer maintains an essentially constant voltage across its terminals over a specified range of the applied voltage. The maximum value of the layer current, I_a, does not lie far from the minimum breakdown current value I_b. It is found that when the layer current exceeds I_a the layer may be damaged.

The breakdown process across the dielectric material

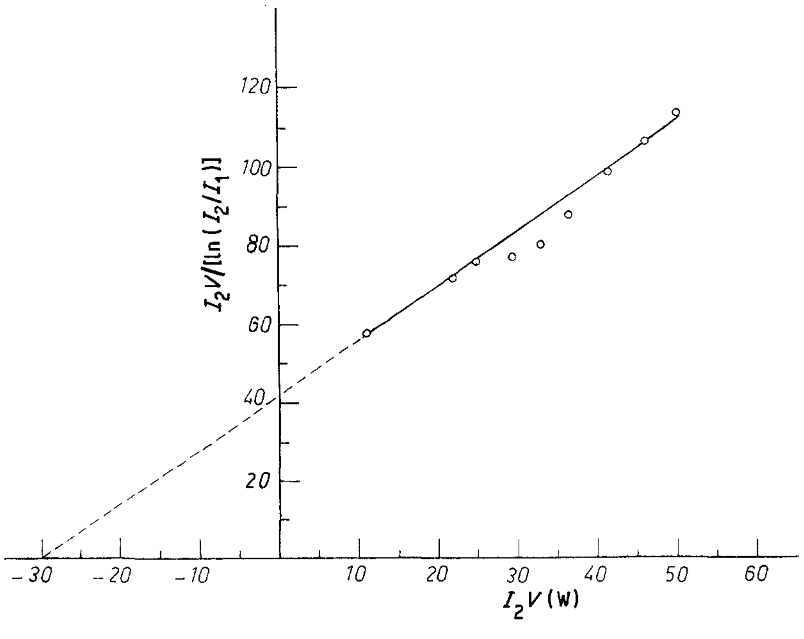

Figure 7 Graph to test Joule heating assumptions, using Newton's law of cooling.

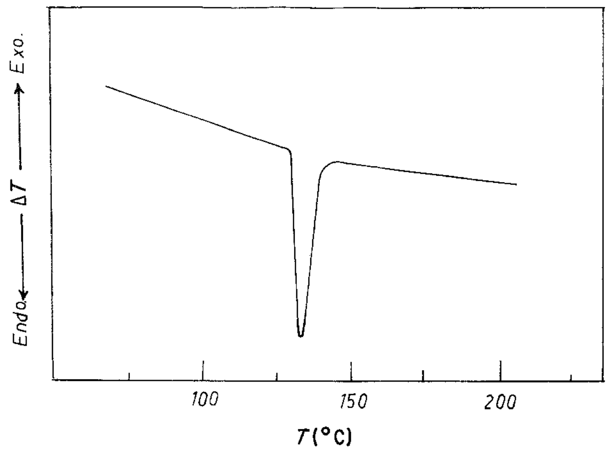

Figure 9 DTA measurements showing a phase transformation in KNO_3.

(KNO_3 single-crystal thin layer) can be explained as follows. When electrodes are separated by a solid insulating material, a surface discharge over the insulator takes place at a lower voltage than across a straightforward gap [23]. At the cathode–insulator junction, free electrons are emitted and these electrons strike the anode or the insulator near the anode, thereby releasing secondary electrons. A positively charged area on the surface of the insulator near the anode moves towards the cathode so that the stress at the cathode eventually increases sufficiently to cause breakdown.

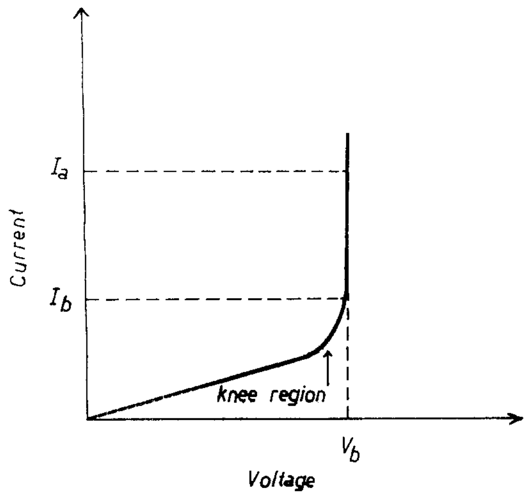

Figure 10 Breakdown characteristics of KNO_3 thin layer.

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