Pak. J. Sci. Ind. Res. 2010 **53** (6) 311-315

Electronic Properties of Au/MgF_2/Au Structures at Different Temperatures

Hossein Ghaforyan[a]*, Hasan Bidadi[b] and Majid Ebrahimzadeh[c]

[a]Payame Noor University (PNU), Miandoab Branch, Miandoab, Iran

[b]Faculty of Physics, University of Tabriz, 51664 Tabriz, Iran

[c]Faculty of Physics, University of Shiraz, Shiraz, Iran

(received February 11, 2010; revised May 25, 2010; accepted June 10, 2010)

Abstract. Investigations of some electronic properties of vacuum evaporated thin film Au/MgF_2/Au structures such as circuiting I_c and emission I_e currents *versus* the applied voltage, electron attenuation lengths MgF_2 layers and the role of the latter layers showed that these devices undergo an electroforming process leading to decrease in resistivity of several orders of magnitude along with a negative resistance region in their *I-V* characteristics. High emission current densities are archived for low applied voltages with the cathodes at or near the room temperature. By decreasing the temperature, both I_c and I_e decreased and at low temperatures the negative resistance region disappears completely. High values of hot electron attenuation lengths in the insulator were obtained and the significance of these high values is described.

Keywords: thin films, cold cathodes, electroforming, Au/MgF_2/Al thin films

Introduction

Thin layers of insulators sandwiched between two metal electrodes exhibit a number of interesting properties. If the insulating layer of a metal-insulator-metal (MIM) device is relatively defect-free and sufficiently thin (< 50Å), it will conduct duo to quantum mechanical tunneling (Simmons, 1963a; Fisher and Giaver, 1961). Once the voltage across the device is sufficiently high so that the conduction band of the insulator at the negatively biased electrode-insulator interface is energetically below the Fermi level of the positively biased electrode then the I-V characteristics obey Fowler-Nordheim characteristics:

$$I = 3.35 \times 10^{17} \left(\frac{V}{t}\right)^2 \left(\frac{A}{\varphi_o}\right) \exp\left\{-\, 0.69t \;\frac{\varphi_o^{\frac{3}{2}}}{V}\right\},$$

where I is the current in mA, V is the voltage in V, t is the thickness in Å, A is the barrier area in m^2 and φ_o is the barrier height in eV (Simmons, 1963b).

The device is then said to be in the field emission regime. At higher insulator thickness, the electric field is not usually high enough for the tunnel current to be measurable and any one of a number of other conduction mechanisms dominate, e.g., Schottky emission (field assisted thermal injection of carriers into the insulator from the metal), Poole-Frenkel emission (field assisted thermal emission of carriers from traps inside the insulator) or space charge limited conduction (Kao and Hwang, 1981).

*Author for correspondence; E-mail: hghaforyan@yahoo.com

If the thickness of the insulating layer of a metal-insulator-metal (MIM) device is in the correct range (100-10000 Å) and the device is placed in a vacuum (< 10-4 mbar) at a high enough temperature (room temperature or above), it undergoes a process known as electroforming when a sufficiently high voltage (greater than the forming threshold voltage) is applied across it. Filaments are thought to be created between the metal electrodes which bridge the insulating gap, and the device is said to be electroformed (or simply formed) (Sharpe and Palmer, 1996). This process can lead to the emission of energetic electrons from the device into the vacuum. Hence, when a bias voltage is applied across an unformed sample, an electroforming process takes place in the dielectric layer and its resistance is decreased. After electroforming, devices in which the dielectric is an oxide or oxide complex generally show a voltage-controlled negative resistance, electroluminescence, electron emission into a vacuum and a possible memory effect. The mechanism of electron transport through thin insulating films sandwiched between metal electrodes has been the subject of a number of theories to explain the current and voltage and related characteristics. One of the models which successfully explain thin layers of insulators sandwiched between two metal electrodes, has been put forward by Dearnaley *et al.* (1970). Their model explains the conduction of field induced metallic filaments in the dielectric matrix sandwiched between the two electrodes thereby providing low resistance paths for the current, and electron emission takes place from the ends of these filaments at the top positive electrode. To study the energy loss mechanism in these samples, the transfer ratio is measured as a function of thickness of the top or the insulator layers which in turn leads to direct measurement of the hot electron attenuation lengths in these layers (Ilyas and Hogarth, 1983). Ghaforyan *et al.* (2008) has shown that, the transfer ratio is defined as the ratio of electron emission current I_e to the circulating current I_c for a given voltage applied across the sandwich. Results of such experiments indicated a strong attenuation of electrons in the top metal electrode and also provided evidence of inelastic interactions of the injected electrons in the top layer (Mordvintsev *et al.,* 1998; Oxley, 1981; Hickmott, 1962). In the present experimental paper, measurements are reported on Au/MgF_2/Au sandwiches and the usual phenomena of electroforming, negative resistance and electron emission into a vacuum are observed for MgF_2 layer.

Materials and Methods

Specimen preparation. Thin insulator layers, used in this experimental work, were prepared by thermal evaporation of magnesium fluoride from a tantalum source in a Balzers 510 coating unit. This unit consisted of evaporation facilities and a vacuum system capable of producing a vacuum of 10^{-6} torr; lower pressures of the order of 10^{-7} torr could be achieved with the addition of a liquid nitrogen trap. A radiant small heater was mounted in the vicinity of the substrates. Masks below the substrates could be rotated by an external rotary seal and a maximum of four different layers could be deposited. For deposition of Au layers, a tungsten crucible with Al_2O_3 layer was used to control the deposition rate and the thickness of the samples; two separate quartz crystal monitors were used. All samples used in this study were prepared by successive evaporation of various layers without breaking the vacuum.

Measurements. All electrical measurements were carried out in a vacuum system capable of maintaining the samples at a pressure of about 5×10^{-6} torr. The I-V characteristics were measured by traditional methods. To measure the temperature dependence of the I-V characteristics and also the electron emission from Au/MgF_2/Au specimens, a small heater made of nichrome wire was used along with a metal cryostat. The heater was placed between the cryostat which could be filled continuously with liquid nitrogen. It was so designed that it would not have an undesirable magnetic influence on the electron emission characteristics. The circuits used are sketched in Fig. 1(a and b). A Kitley G10 electrometer was used for measurement of the emission currents with the accuracy of 10^{-15} A.

A constant power supply unit was used to feed the heater. By the judicious use of cryostat and heater, the required temperatures could be obtained. Device temperatures were recorded

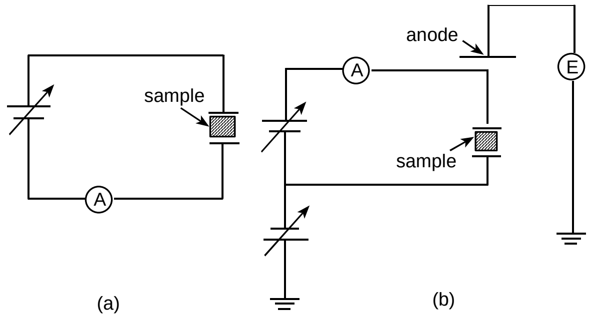

Fig. 1. Circuits for measurement of **(a)** circulating currents **(b)** emission currents.

on a copper constantan thermocouple at the surface of the substrate. Electron emissions into vacuum were collected at a copper anode (2.5 cm × 7.5 cm × 0.1 cm) placed at a distance of 1.0 cm from the specimens; anode was kept at a potential of 100 volt with respect to earth. As shown in Fig. 2 (a and b), for one evaporation procedure, six specimens were prepared on each substrate.

Attenuation length measurements. Measurement of the relative numbers of electrons emitted through the thin top layer as a function of layer thickness yields values of the hot electron

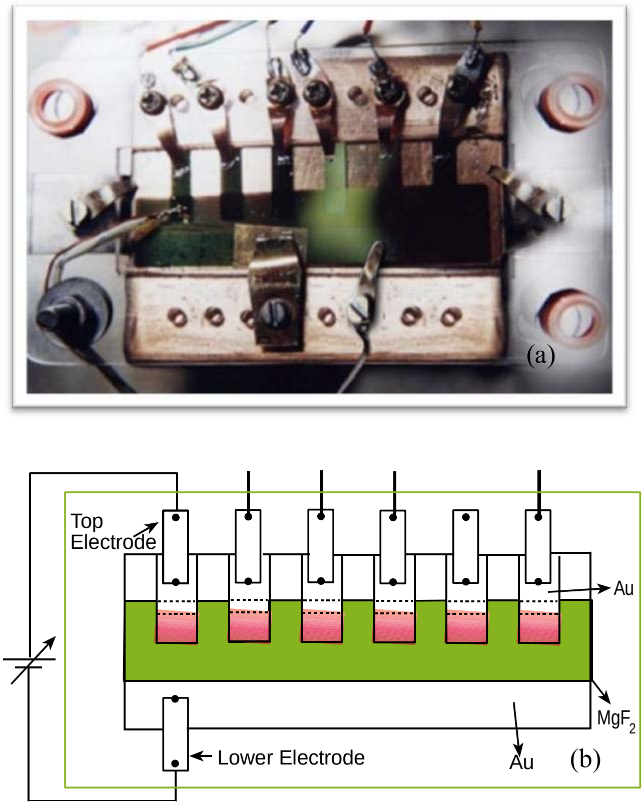

Fig. 2(a) View of the set-up implemented in this work **(b)** Schematic view of the set-up.

attenuation length directly. To determine λ_{MgF_2} experimentally, samples with constant top electrode thickness but with insulator layers of different thickness were prepared.

Results and Discussion

Electrical characteristics. The conductivity of the samples increased by several orders of magnitude when they were formed at the room temperature in vacuum. As the voltage across the sample was increased, critical voltage was reached at which a sharp increase in current through the insulator layer occured. On lowering the current through the sample, a pronounced negative resistance region occured. On raising the voltage to successively higher values, the current voltage characteristic continued to show a negative resistance region for both the increasing and decreasing voltage. Currents through the MgF_2 layer were noisy and highly erratic during the first time that a voltage range was covered. However, on successive tracing of a characteristic, the currents were much less erratic. The electroforming process consisted of cycling the voltage, applied across the sandwich between 0 and 14 V with top electrode positively biased. The voltage required for the forming increased with the increase or dereasing insulator thickness and it was found possible to form sandwiches electrically in which the insulator thickness was 1.5 μm. The forming voltage increased with the decreasing temperature. The voltage at which the maximum peak current occured varied between 2 and 3 V for diodes having different material (Au and Cu) and seemed to increase only very slightly with the increase or decrease thickness of the insulator layer. The onset of electron emission into a vacuum occurred for the same applied voltage as that at which the voltage controlled negative resistance (VCNR) occurred. A typical curve illustrating the circulating and emission currents in a $Au/MgF_2/Au$ sandwich is shown in Fig. 3 and 4.

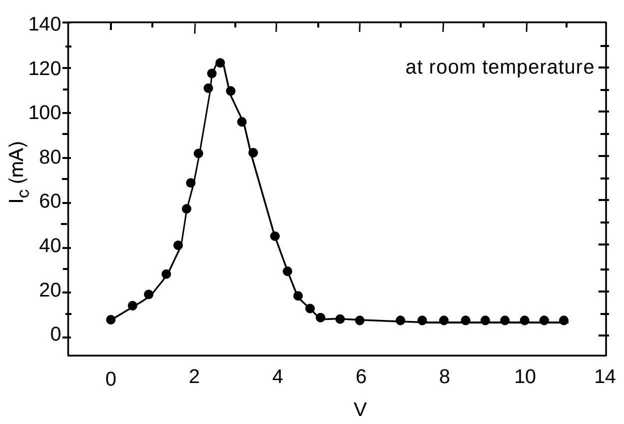

Fig. 3. Variations of circulating current *vs* the applied voltage for $Au/MgF_2/Au$ sample (at room temp.)

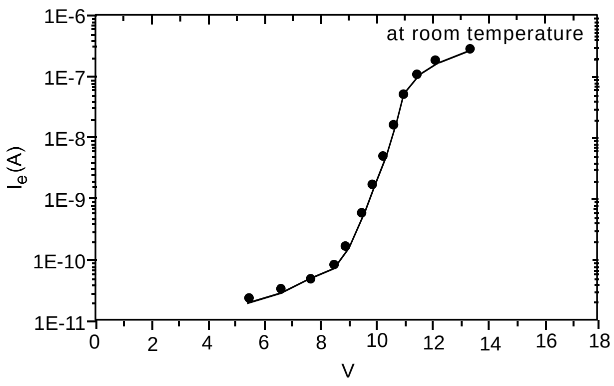

Fig. 4. Variations of emission current *vs* the applied voltage (at room temp.)

Temperature dependence of electrical characteristics. The electrical conductivity and the shape of the I-V characteristics were very sensitive function of temperature after the sandwiches had been electrically formed and significant conductivity had been established at room temperature (Gravano and Gould, 1992). As the temperature was lowered to 238 K, the maximum circulating current and the general current level decreased gradually. The current including the peak value increased considerably as the temperature was raised to 333 K. Fig. 5 shows the temperature dependence of specimen conductivity at three different temperatures for a freshly made $Au/MgF_2/Au$ sample. Electron emission into a vacuum was observed as conductivity developed in the insulator and this emission current was also temperature dependent.

Attenuation length measurements. Fig. 6 shows log α = log I_e / I_c as a function of thickness of the MgF_2 layers for different values of applied voltage. The slopes of the lines are steeper for electrons emitted at lower applied voltages and indicate that the electrons of lower energy are more heavily

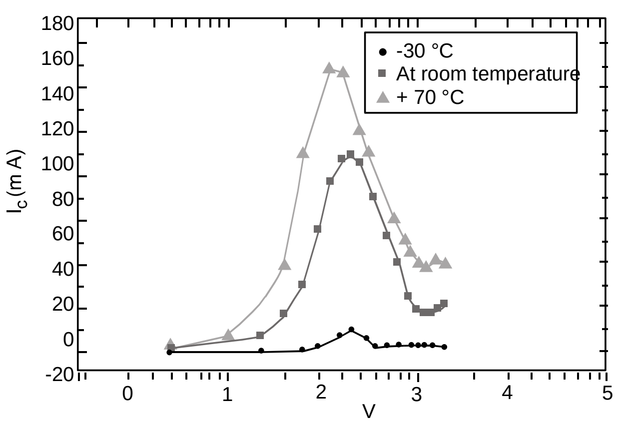

Fig. 5. Variations of circulating current *vs* the applied voltage at three different temperatures.

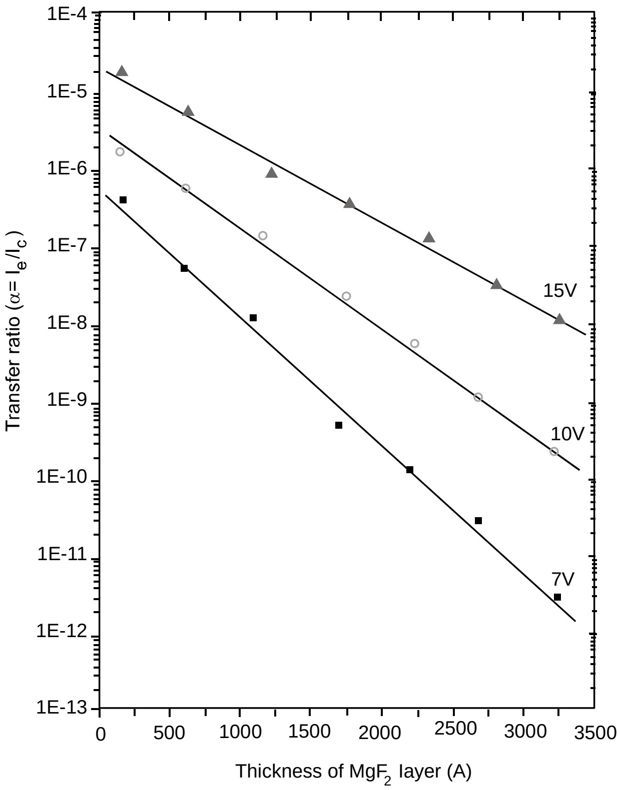

Fig. 6. Variations of transfer ratio *vs* ($\alpha = I_e / I_c$) the thickness of MgF_2 layers.

attenuated than those of higher energies. Experimental evidence for the energy loss is provided by the fact that, in general, the transmission ratio increased with the increase in the applied voltage. From the slopes of the graphs (Fig .4), an attenuation length was derived for each value of the applied voltage in the range of 6-15 volts. The values of λ_{MgF_2} were approximately constant (1200Å) in the range 7-13 V and then inceased with the increase of voltage to a value of 1800Å at 15 V applied.

Conclusion

Reasonably good electron emission properties were found and hot electron attenuation lengths were determined. The general electrical properties of Au/MgF_2/Au sandwich structure were similar to those of the other metal-insulator-metal structures and point to the existence of a barrier layer near the cathode which can cause free electrons to get accelerated to very high energies, sufficient to penetrate the remainder of the insulator, the top metal electrode and emitted into vacuum. Increase in circulating and emission currents at a given bias voltage observed at high temperature is expected since more thermal energy is available for transfer to the total energy of the electrons. The reduction density observed was of the order of 1×10^{-6} A. cm^2. One of the more successful models of these phenomena is the filamentary model (Dearnaley *et al.*, 1970), in which it is assumed that the electroforming process establishes a population of ohmic filaments within the insulating matrix, which spans the metal contacts. The VCNR behaviour results from progressive cessation of conduction in individual filament owing to Joule heating effects. By postulating plausible probability distributions of filament resistances, Oxley (1980) obtained typical I-V charac-teristics which accorded with our experiment.The polyfilamentary model of gold seems to indicate that a practical emission level of emission current was achieved. The circulating current initially increasesed with voltage faster than expected according to Ohm's law, following typically either an $I \alpha I_0 \sinh(V/V_0)$ or an $1 \alpha I_0 (V/V_0)^2$ dependence, where I is the current through the sample, V is the applied voltage, and I_0 and V_0 are constants (Dearnaley *et al.*, 1970). The conducting filaments were initially characterised in terms of a probability density distribution of filament resistances P(ρ) which was triangular, where P represents the filament resistance. Later work on the form of the function P(ρ) suggested that a parabolic distribution equally well described the experimental results (Gould, 1979), while it was also argued that a normal distribution of filament resistances was more fundamental (Ray and Hogarth, 1985), and that the earlier suggested distributions were merely approximation to this. More recently the present authors have suggested that there is not *a priori* reason for suggesting that the filament resistance is normally distributed, and that more basic attributes such as the filament radius (Gravano and Gould, 1992) or cross-sectional area (Gravano and Gould, 1994) may be normally distributed. Indeed, both of these assumptions have been shown to yield computed characteristics in accordance with our experiment.

The transport and energy loss mechanism of hot electrons in metal films have been extensively studied both theoretically and experimentally (Carey and Silva, 2001). To study the energy loss mechanism in these samples, the transfer ratio α is measured as a function of the thickness of top or the insulator layers. The transfer or transmission ratio is defined as the ratio of electron emission current I_e to the circulating current I_c for a given voltage, applied across the sandwich. Results of such experiments indicated a strong attenuation of electrons in the top metal and also provided evidence of inelastic interaction of the injected electrons in the top layer. Thurstans and Oxley (2002), described a new model of the electroformed metal-insulator-metal structure which explained its various properties including electron emission, electroluminescence, memory effects and, for the first time, a complete account of differential negative resistance (Silva *et al.*, 2001). Measurement of the relative numbers of electron, transmitted through the thin layer as a function of thickness of the layer, yields direct measurement of the attention length. By extending the range of the measured insulator thicknesses, it is possible to find out the values of attenuation length of hot electrons in the insulator.

References

Carey, J.D., Silva, S.R.P. 2001. Electron field emission from amorphous semiconductors. *Solid State Electronics*, **45:** 1017-1020.

Dearnaley, G., Stoneham, A.M., Morgan, D.V. 1970. Electrical phenomena in amorphous oxide films. *Reports on Progress in Phys*ics, **33:** 1129-1192.

Fisher, J.C., Giaever, I. 1961. Tunneling through thin insulating layers. *Journal of Applied Physics,* **32:** 172-177.

Ghaforyan, H., Bidadi, H., Ebrahimzadeh, M. 2008. Electronic properties of Au/CeF_3/Au structures at different temperatures. *International Journal of Pure and Applied Physics,* **4:** 113-118.

Gould, R.D. 1979. Relationship of the current-voltage characteristics to the distribution of filament resistances in electroformed MIM structures. *Thin Solid Films,* **57:** 33-38.

Gravano, S., Gould, R.D. 1994. Derivation of the current-voltage characteristics in filamentary conductors as a function of the distribution of filament cross-sectional areas. *Thin Solid Films*, **248:** 263-266.

Gravano, S., Gould, R.D . 1992. Calculations of the current-voltage characteristics of electroformed MIM structures, assuming a normal distribution of filamentary radii. *International Journal of Electronics*, **73:** 837-839.

Hickmott, T.W. 1962. Low-frequency negative resistance in thin anodic oxide films. *Journal of Applied Physics*, **33:** 2669-2682.

Ilyas, M., Hogarth, C.A. 1983. The optical absorption edge of amorphous thin films of borosilicate glass. *Journal of Material Science Letters,* **2:** 535-537.

Kao, V.K.C., Hwang, W. 1981. *Electrical Transport in Solids with Particular Reference to Organic Semiconductors,* 663 pp., Pergamon Press, Oxford, UK.

Mordvintsev, V., Kudryavtsev, S., Levin, V. 1998. Aspects of the self organization of carbonaceous conducting nanostructures during electroforming of a metal-insulator -metal open sandwich structure with a nanometer-size insulating gap. *Technical Physics*, **43:** 1350-1357.

Oxley, D.P. 1981. Memory effects in oxide films. In: *Oxides and Oxide*, A. K., Vijh (ed.), vol. **6**, pp. 251-325, Marcel Dekker, New York, USA.

Ray, A.K., Hogarth, C.A. 1985. A note on the normal distribution of filamentary resistances in formed metal/ insulator/metal devices. *Thin Solid Films*, **127:** 69-76.

Sharpe, R.G., Palmer, R.E. 1996. Evidence for field emission in electroformed metal-insulator-metal devices. *Thin Solid Films*, **288:** 164-170.

Silva, S.R.P., Carey, J.D., Poa, C.H., Shannon, J.M. 2001. Conditioning of amorphous cathodes via current stressing. In: *Proceedings from the First International Symposium on Cold Cathodes*, M. Cahay, K.L. Jensen, P.D. Mumford, J. Yater, R. A. Murphy, D. Temple and V.J. Kapoor (eds.), vol. **2000-28**, pp. 226-246. The Electrochemical Society.

Simmons, J. G. 1963a. Generalized formula for the electric tunnel effect between similar electrodes separated by a thin insulating film. *Journal of Applied Physics,* **34:** 1793-1804.

Simmons, J.G. 1963b. Electric tunnel effect between dissimilar electrodes separated by a thin insulating film. *Journal of Applied Physics*, **34:** 2581-2590.

Thurstans, R.E., Oxley, D.P. 2002. The electroformed metal-insulator-metal structure: a comprehensive model. *Journal of Physics, D: Applied Physics*, **35:** 802-809.