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Physics and Technology of Metal-Insulator-Metal thin film structures used as planar electron emitters

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PHYSICS AND TECHNOLOGY OF METAL-INSULATOR-METAL THIN FILM STRUCTURES
USED AS PLANAR ELECTRON EMITTERS**

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Abstract:

Physics and technology of Metal-Insulator-Metal thin film structures used as planar electron emitters.

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Electron projector for studies of properties of M-I-M plane cathodes.

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The electron projector has been designed for observation of electron emission pattern from MIM tunnel planar cathodes, and measurements of their electronical properties *in-situ*. Since the performance of the MIM cathodes can be influenced by various kinds of impurities, the ultra-high vacuum construction with oil-free pumps was used.

1. Introduction

The forming processes in MIM sandwich structures limit their lifetime, especially if such a structure is to be used as a carrier of pattern information. These processes are undesirable for an electron imaging system (Delong and Kolarík 1989, Hladil et al. 1991).

To improve the preparation technology it is necessary to study these processes by means of a combination of some kind of spectroscopy with emission pattern observation (Drstíčka and Pavelka 1990, Pavelka et al. 1991).

The electron projector is designed for this purpose. It facilitates electron emission pattern observation, measurement of C-V characteristics, impedance analysis, and other *in-situ* measurements up to 13 MHz within the cathode temperature range 77 to 500 K, and at the pressure down to 6×10^{-10} Torr *in-situ*.

2. Description

The emission pattern is imaged onto a scintillation screen using homogeneous parallel electric and magnetic fields. The relation between the distance of the image plane from the cathode d, the accelerating voltage U across the distance, and the magnetic flux density B is as follows:

$$2 \cdot e^2 m_e U$$

$$d^2 =$$

$$e B^2$$

where m_e is the electron mass, e is the elementary charge.

The basic values of these quantities chosen in the projector are:

$$d = 12 \text{ mm}, U = 5 \text{ kV}, B = 0.063 \text{ T}.$$

The projector body, with the scintillation screen, and the viewport, the cryostat sample holder, the vacuum system, and the frame form a compact desktop unit. The arrangement is shown in Fig.1.

The group of power supplies, and the Dewar vessel with the feed pipe for the cryostat sample holder are assembled separately. The supplies are used for: the accelerating voltage, the tunneling current through the MIM cathode, the current for magnetic field excitation, the voltage for the sputter-ion pump, and the heating jackets for degassing process.

The heart of the projector is a flat cylinder chamber. It is made of stainless steel, except of two tubes which are made of magnetically soft stainless steel - permalloy - and serve as parts of magnetic polepieces.

Near one of the Permalloy tubes, in a distance of 6 mm off the main symmetry plane, there is a scintillation screen holder. The voltage of 5 kV necessary for a scintillation image observation is connected to the holder via a feedthrough placed on one of the side flanges. The image is observable via the viewport on the flange contiguous with the screen holder. A monocrystalline yttrium-aluminum garnet (YAG) plate serves as scintillation screen.

There are two coils and iron plates put on the chamber. The arrangement facilitates an easy removal of the coils during the degassing of the apparatus. The coil together with the iron (behanit) nickel-coated plate and mantle forms a compact block. The iron plates together with the permalloy tubes form magnetic polepieces.

The body of the LN₂ flow cryostat is terminated in a head, which is equipped for a placement of the cathode. From four coaxial feedthroughs with floating shields (SMC type) lead the measurement terminals. The cryostat is connected to the projector body by means of an extension piece. The cryostat is connected to an LN₂ container by means of a feed pipe, to a pump controlling the flow of the coolant, and to a controller for an adjustment of the temperature of the cryostat head with the cathode.

The cryostat consists of two heat exchangers, the first heat exchanger (the head) carrying a sapphire plate on which the cathode can be placed (max. diam. 22 mm), and the 2nd heat exchanger serving for the compensation of heating of measuring terminals.

The pumping system consists of two rough vacuum adsorption pumps, two angle valves, a bakable "T" valve, a venting valve, a Pirani gauge, an ion gauge, and a sputter-ion pump.

The projector has been manufactured by Delong Instruments.

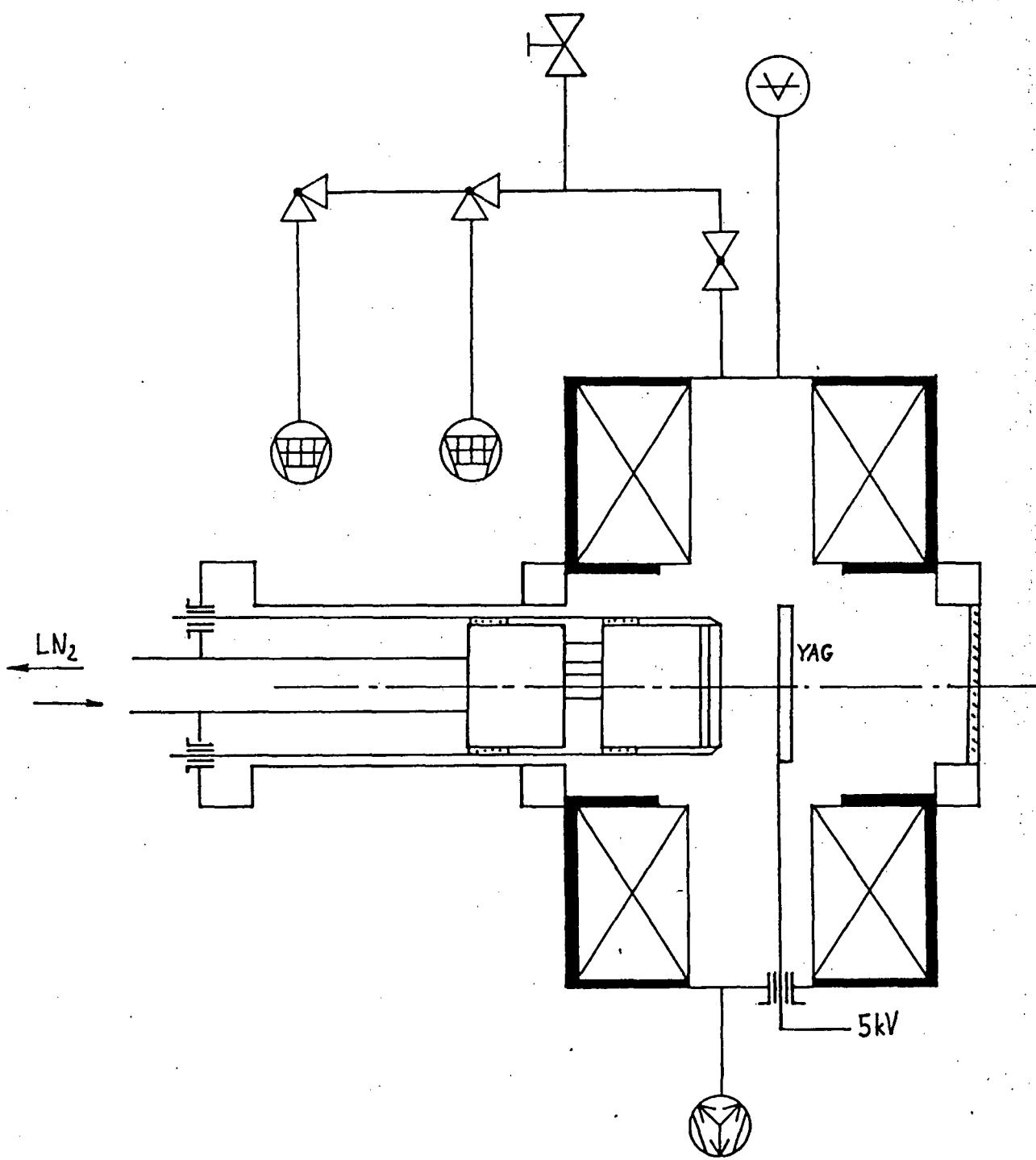
The first measurements using this equipment have been made in Institute of Mathematics and Physics, Roskilde University Center, Denmark (Olsen et al. 1991).

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Fig. 1. Scheme of the electron projector

Running headline: Electron projector.



M-I-M cathodes, their application to electron lithography
and characteristics of the emitted electrons

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This paper gives a brief account of the possibility of applying MIM (metal-insulator-metal) cathodes to projection electron lithography (a 1:1 scale), and also a description of the measurement of the energy distribution and angular distribution characteristics of the electrons emitted from MIM cathodes.

1. Introduction

For the envisaged application of MIM cathodes in 1:1 electron -projection lithography /1/ it is necessary to measure parameters so as to enable an objective evaluation of the properties and the technology of preparation of MIM structures.

2. Working principle

Fig. 1 shows schematically a MIM tunnel cathode during exposure in an electron-projection lithograph. On an Si substrate of 50 mm in diameter with an SiO_2 insulation layer there is a sandwich made up of layers of Al, Al_2O_3 , Au. Current I_d is set to such a value that in areas where the Al_2O_3 layer is 11 nm thick there is a tunnel current flowing, whose density is up to 100 mA/cm^2 . In consequence of the steep dependence of emission current I_e on the field strength in the dielectric, (Fig. 2) emission from areas with thicker (18 nm) dielectric is practically zero. Obtaining a high degree of contrast and creating submicron structures on areas of up to 10 cm^2 of the MIM cathode is no longer a technological problem.

The emission current has a density of about $100 \mu\text{A/cm}^2$. The electrons emitted from the MIM cathode are accelerated and focussed by homogeneous parallel electric and magnetic fields on the surface of exposed wafer. In this arrangement we obtain a 1 : 1 projection from the cathode surface onto the wafer. The exposure time for an area of up to 10 cm^2 is about 0.1 s.

3. Results of measuring the characteristics of tunnel MIM cathodes

The volt-ampere, emission and energy characteristics were measured in an electron projection stepper /1/ as shown in Fig. 1, with the difference that the wafer was replaced by a flat carbon anode and the cathode-to-anode distance was reduced to 1 mm. Measuring U_d , I_d , U_a , I_e , I_a was controlled from a HP 2486A measuring central. The size of the emission area of the MIM cathode was 2.25 cm^2 .

Fig. 2 gives a typical volt-ampere characteristic of an MIM cathode, where U_d is the voltage between the aluminium and the gold layers, I_d is the current flowing through the dielectric layer, and I_e is the emission current. The steepness of the characteristics is somewhat reduced due to the resistance of the gold surface layer. The energy distribution of emitted electrons was measured for $I_d = 10$ or 100 mA , which corresponds to a current density of 4.44 and 44.4 mA/cm^2 respectively. By differentiating the anode current measured with respect to anode voltage we obtained a dI_a/dU_a curve characterizing the energy distribution of the electrons emitted. The width of energy spectrum at half the height is 0.66 eV for $I_d = 10 \text{ mA}$ and 0.85 eV for $I_d = 100 \text{ mA}$. The shift in U_r and the reduced slope of the dI_a/dU_a curve, which are apparent especially for larger I_d , are due to the voltage drop in the surface gold layer, whose resistance per square is about 25 ohms . When processing the measurement results a correction of 0.35 V was included which was given by the difference in the work functions

of the Au surface of the cathode (4.71 eV) and the carbon anode (4.36 eV). There is good agreement between the values measured and theoretical conclusions /2/.

To determine the angular distribution characteristics a line motive was created on the MIM cathode surface by means of an electron lithograph. The motive was made up of 12 lines 5 μ m thick, see Fig. 5b. Instead of the wafer from the set-up in Fig. 1 a monocrystalline YAG-Ce screen. An optical system was used to observe and photograph the image formed by electrons on the YAG-Ce screen. The distance between the cathode and the screen surface was increased to 15 mm, voltage U_a was 5 kV. The upper part of Fig. 5b shows the picture of the line motive in focussed condition with magnetic field B. The middle part of the Figure gives the same motive but in not focussed condition, when the magnetic field is off and electrons which are not perpendicular to the cathode move along parabolic trajectories, as can be seen in Fig. 5a. From the optically measured widening of the lines it follows that the tangential components acquire values of up to 0.68 eV, which value is higher than anticipated on the basis of the theory and the resolution observed in an emission electron microscope /3/. This disagreement can be attributed especially to the unevenness of the vacuum-deposited Al layer , which is also copied by Al_2O_3 layer, or to the interactions of the electrons in the Au layer. It will be necessary to repeat the measurement with smooth layers formed e.g. by epitaxy technology. To measure exactly the current flow in the defocussed line motive we prepare a sliding slot probe.

4. Conclusions

Cold tunnel MIM cathodes have several good properties which offer a wide range of application in projection electron lithography. Their advantages are: large working areas, emitting at room temperature, high degree of contrast, good resolution and, above all, no need for any costly attachment, such as a synchrotron or soft X ray laser.

The above measurements of energy distribution of emitted electrons and tangential components will be used as input data for the calculation of the resolution power of electron lithographic systems with MIM cathodes.

Acknowledgements

The authors wish to express their thanks to RNDr.M.Lýčka for excellent cooperation when processing the results measured.

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Running headline:

K.Hladil et al.: M-I-M cathodes and characteristics of the emitted electrons.

Fig. 1: Principle of 1:1 projection of MIM cathode as a mask

- 1 - MIM cathode
- 2 - emitted electrons
- 3 - wafer

Fig. 2: V-A and emission characteristics of a MIM cathode

Fig. 3: Energy distribution of emitted electrons ($I_d = 10 \text{ mA}$)

Fig. 4: Energy distribution of emitted electrons ($I_d = 100 \text{ mA}$)

Fig. 5: Measurement of tangential energy components of emitted electrons

- a - principle of measurement

- 1 - MIM cathode
 - 2 - trajectories in homogeneous electric field
 - 3 - fluorescent screen
 - 4 - camera

- b - photographies of focused and not focused images of line motive

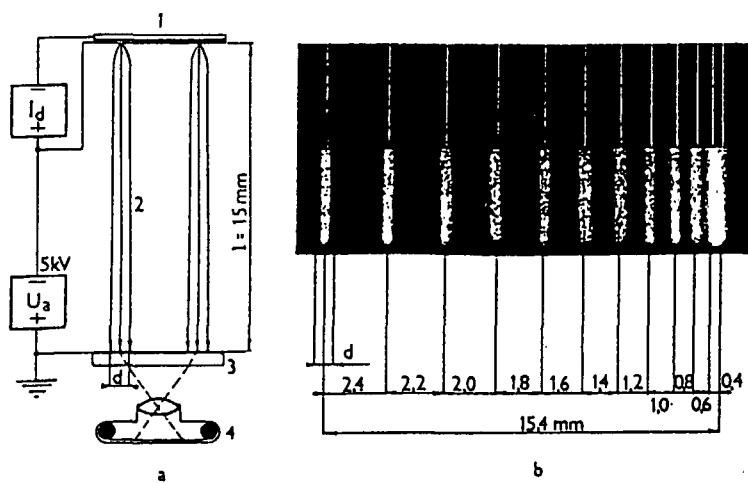
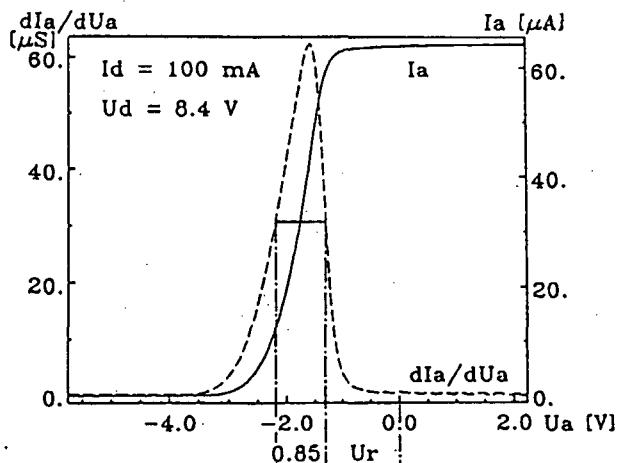
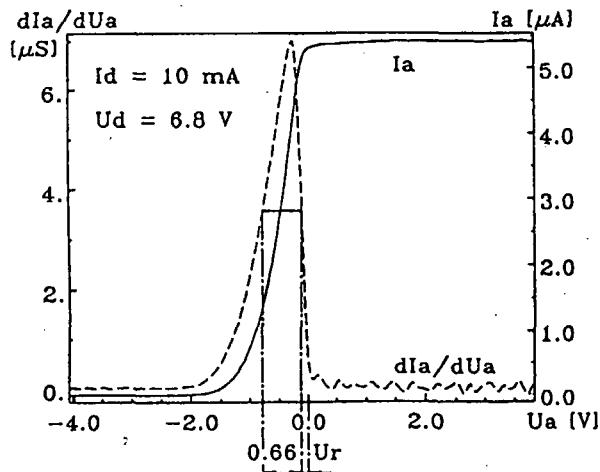
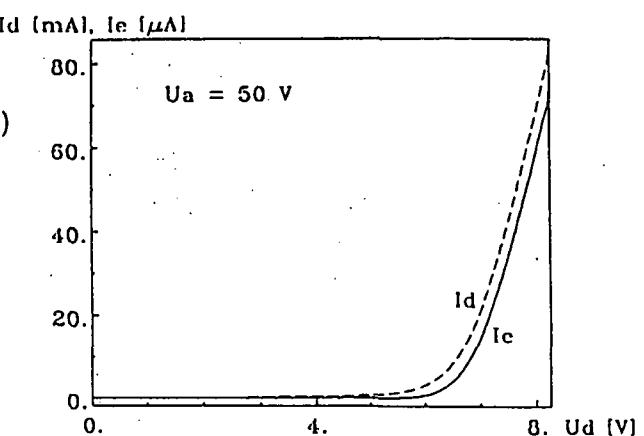
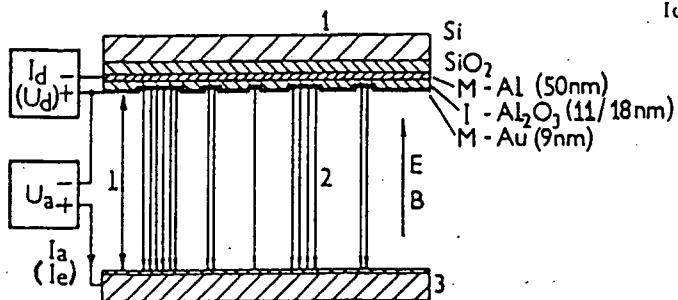


Fig. 5

ELECTRICAL PROPERTIES AND EMISSION PATTERN FROM Al-Al₂O₃-Au
METAL-INSULATOR-METAL TUNNEL CATHODES.

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Abstract:

The electrical properties and the emission pattern from Al-Al₂O₃-Au Metal-Insulator-Metal (MIM) sandwich structures have been investigated. Under certain conditions the MIMs exhibit Negative Differential Resistance (NDR), in which case the electron emission is non-homogeneous.

1. Introduction

Al-Al₂O₃-Au Metal-Insulator-Metal (MIM) tunnel cathodes are to be used in the new generation of multibeam electron lithographers as 1:1 projection electron steppers (Delong and Kolarik 1989). The main advantages are high electron emission density, submicron resolution of the 1:1 projection and the possibility of parallel rather than sequential exposure.

The main problem is the lifetime of the cathode, i.e. the time in which the electron emission is homogeneous. The non-homogeneous emission from a destroyed cathode will be seen as spotty sparks on a homogeneous background. Observation by optical microscope shows pinholes in the surface and transmission electron microscopy reveals holes in the Al₂O₃ layer (up to 600 nm) (Drsticka and Pavelka 1990).

2. Results

The MIMs have been studied in an electron projector where the emission pattern from a MIM cathode held at the tip of a liq. nitrogen flow cryostat can be observed by means of an Ytrium-Aluminium-Garnet scintillator by accelerating the emitted electrons and projecting the emitting surface (1:1 projection in parallel electric and magnetic fields) on the surface of the scintillator (Drsticka et al. 1991).

Previous observations and study of electron emission have been

performed at and above the room temperature, and a non-homogeneous emitting cathode exhibits in this case a region of Voltage-Controlled Negative Resistance (VCNR) in the I-U characteristics, where I is the current between the electrodes and U is the external applied voltage.

Two kinds of non-homogeneities in the electron emission have been observed; stable spots of higher intensity, caused probably by defects in production, and unstable (subsecond) temperature dependent sparks which begin when the VCNR peak starts to rise and which are visible only within the VCNR region. The number of stable spots of higher intensity changes from sample to sample and some haven't any at all.

If there is no region of VCNR, the electron emission will be homogeneous (apart from possible stable defects). The non-homogeneous emission must be therefore connected with the phenomena of Negative Differentiel Resistance (NDR).

It is possible to repeat the temperature and VCNR cycles several times using the same MIM, thereby changing the electron emission pattern from being more homogeneous to non-homogeneous and back. The pinholes in the surface and the holes in the insulator layer, created presumably in the state of non-homogeneous emission, are however believed to be still present, even though the MIM is emitting homogeneuosly on an optical length scale.

3. Conclusion

Although the nature of the NDR behavior has been discussed in the litterature (e.g. Pagnia and Sotnik 1988) its nature has still to be considered as unknown at present.

The preparation technology of the MIMs (Drstickle and Pavelka 1990) are believed to cause the stable defects and steps are taken to exchange it by molecular beam epitaxial growth of the MIM layers.

The connection between non-homogeneous electron emission and the existance of NDR has been confirmed, but the nature of NDR is not yet understood. Electron emission pattern should be observed at higher resolution, and other preparation technologies should be tried in order to prevent NDR and eventual destruction of the cathodes.

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M-I-M tunnel cathodes for the 1:1 electron stepper -
technology of preparation and structure description.

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A metal-insulator-metal tunnel structures have been used for an image generation in the 1:1 electron stepper. The conditions of performance of such a cathode place high demands on physical properties of all the components of the system. The Al-Al₂O₃-Au sandwich has proved to be the most simple and reliable. The properties of the system are strongly affected by the technology of preparation of the layers. To improve the preparation technology, different deposition techniques were used for the system creation and the processes going on in the cathode during operation have been analyzed.

1. INTRODUCTION

To find a productive lithographic system which is able to create submicron structures (0.2μ) on a chip of a large size (30 x 30 mm) at a reasonable price, that's one of the main tasks in the microfabrication at present. The 1:1 electron stepper (De long and Kolařík 1989) appears to be a very promising projection system which could be able to satisfy the demands mentioned above. In spite of many advantages of the system, the main problem which hasn't been satisfactorily solved, is relatively short lifetime and low reproducibility of

preparation of the MIM tunnel cathode which has been used in this device.

2. DISCUSSION

Different materials deposited by means of different methods were used for the MIM structure creation. Evaporated Ta/anodic Ta_2O_5 /evap. Au and similarly Zr/Zr $_2O_5$ /Au systems doesn't show appropriate tunneling characteristics, the oxides behave more like semiconductors, they seem to be contaminated by oxides of W or Mo (Ta, Zr evaporated from W or Mo boat). In the sandwich of Si single-crystal/anodic SiO_2 /evap. Au the work function from Si to SiO_2 is relatively high (3.2 eV), and stronger electric field which has to be put on the cathode (to obtain sufficient emission current) causes the structure destruction in short time.

Concerning the lifetime, the best results have been obtained with the combination of evaporated Al, anodic Al_2O_3 and evaporated Au, prepared on SiO_2/Si substrate. This system is able to produce an electron image with sufficient emission current density for approx. 1-2 hours continuously. After this time the cathode resistance decreases, the emission current drops down, a point emission (sparks) appears in the emitting area, and finally the emission from this area disappears.

It has been shown (Olsen et al. 1992) the undesirable point emission is a phenomenon accompanying electroforming processes going on in the dielectric layer (Drštíčka and Pavelka 1990, Pagnia and Sotnik 1988). Hydrocarbons, incorporated into the insulator probably during the structure preparation, create conductive filaments across this layer. During the operation of the cathode the filaments break and it can result in the point emission effect. This process is supposed to go on in places where some point defects, inhomogeneities or impurities occur in the sandwich. In such a place higher gradient of the electric field occurs, and then strong local diffusion of ions can cause a dielectric breakdown or conditions for filament creation.

The Al/Al₂O₃/Au sandwich isn't completely smooth and planar ~~(Fig. 1)~~. The Al surface is rough, formed by microcrystals and anodic Al₂O₃ is copying the surface profile, fortunately with uniform thickness (Kienzer et al. 1987). Partially coalesced Au islands create the upper layer. Such an arrangement is not ideal, the grain boundaries can happen potential sources of breakdown or filament creation.

3. CONCLUSION

The improvement of the properties of the MIM tunnel cathode (lifetime, reproducibility) requires further research. It will be necessary to apply other materials or deposition methods. Using materials in the single-crystal form, smooth and homogeneous MIM structure without inhomogeneities could be obtained. It might prevent the occurrence of the undesirable phenomena which result in the cathode destruction.

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ELECTRICAL IMPEDANCE SPECTROSCOPY AND ELECTRICAL RESPONSE IN
METAL/NONMETAL SYSTEMS

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Abstract

A new analysis of the electrical response of a macroscopic solid state system to an arbitrary electrical voltage input is presented. The results indicate that with a small, sinusoidal voltage input within the classical frequency range ($\omega < 10^{10}$ Hz), the electrical response in spatially homogeneous systems mirrors first of all the static, spatial distribution of the mobile charge carrier density through the local electrical conductivity relaxation time. This enables any system, inclusive interface and electrical contact regions to be modelled by a simple, passive R,C electrical network, where each of the network elements has a very direct physical meaning. Ultra pure, single crystal silicon, pure polycrystalline silicon and Al-Al₂O₃-Au Metal-Insulator-Metal (MIM) structure are used as illustrative examples.

1. Electrical response at classical frequencies

Mathematical formulation.

The macroscopic electrical response of a medium (characterised by a dielectric constant ϵ and the electrical mobility μ) at classical frequencies is described fully by classical electrodynamics (two Maxwell equations), the constitutive equation defining the total local current, the initial condition for the mobile charge carrier density distribution and by the boundary condition defining the charge transport across the boundaries. Under these conditions the dielectric constant ϵ and the electric mobility μ of the mobile electrical charges can be both considered as space-time independent constants.

In one dimension and when both types of the mobile charge carriers (electrons and holes) contribute to the transport, the defining equations can be re-cast into a set of two, coupled, non-linear parabolic equations for each type of the mobile charge carrier particle density:

$$\frac{\partial m_e(x,t)}{\partial t} = \mu_e m_e(x,t) \cdot \frac{\partial \vec{E}(x,t)}{\partial x} + \mu_e \vec{E}(x,t) \frac{\partial m_e(x,t)}{\partial x} + \frac{\mu_e kT}{|e|} \cdot \frac{\partial^2 m_e(x,t)}{\partial x^2} \quad (1)$$

$$\frac{\partial m_h(x,t)}{\partial t} = -\mu_h m_h(x,t) \frac{\partial \vec{E}(x,t)}{\partial x} - \mu_h \vec{E}(x,t) \frac{\partial m_h(x,t)}{\partial x} + \frac{\mu_h kT}{|e|} \cdot \frac{\partial^2 m_h(x,t)}{\partial x^2}$$

where

$$\text{div } \vec{E}(x,t) = \rho(x,t) / \epsilon \quad (2)$$

Here μ_e and μ_h are the electrical mobilities of the respective charge types and ϵ is the dielectric constant.

$n_e(x,t)$, $n_h(x,t)$, $\vec{E}(x,t)$ and $\rho(x,t)$ are the particle densities of the respective mobile charge carriers, the local electrical field and the local, total charge density respectively.

2. Results

2.1. Dynamical solution.

Equations (1) and (2), together with the appropriate initial and boundary conditions for $n_e(x,t)$ and $n_h(x,t)$ then determine the space-time evolution of these mobile charge carrier densities and therefore determine also the electrical current response to a given applied voltage input, thereby defining the electrical impedance of the system.

2.2. Long time, static limit.

For times $t \gg 0$ and with no external applied field, the set of equations (1) and (2) leads to the formation of the mobile charge carrier depleted regions near the surfaces if the boundary condition reflects the finite difference in the electro-chemical potential across the boundary at time $t = 0$. In this case the space-time evolution of $n_e(x,t)$ and $n_h(x,t)$ will approach the equilibrium, time independent distributions as the time goes to "infinity" $t \rightarrow \infty$. These distributions are also the solutions to a static Poisson equation (equation (2)) to which the problem reduces in this long time, static limit.

2.3. Small signal approximation and the static R,C network solution to the problem of the electric response.

One of the major results of the present dynamical analysis of the electrical response in solids is concerned with a small signal approximation. When the external applied voltage is suffi-

ciently small ($e \cdot V_{ext}^{MAX} \approx kT$), the equilibrium, spatially non-homogeneous distribution of the mobile charge carrier densities will not be disturbed by the applied external field and it can be shown that the electrical response of the entire system under these conditions is identical to a response from simple, parallel R,C electrical elements, connected in series.

Contrary to the usual passive R,C network models of various junctions, interfaces and semiconductor-insulator-metal structures, the electrical elements in the present static R,C network have a very direct physical meaning and are all interrelated.

According to the result of the present analysis, the sample is simply devided into a number of volume elements (the actual number dependinig on the required precision with which the electrical response is required), each volume element v_i being characterised by its electrical resistance R_i (in-phase component of the response; dissipation of energy through finite mobility) and by its geometrical capacitance C_i (out-of phase component of the response; non-dissipative polarisation through finite dielectric constant ϵ).

The total electrical impedance of the system $\tilde{Z}(\omega)$ is then the sum of the impedances of the individual volume elements and it becomes frequency dependent when the local electrical conductivity relaxation times among the respective volume elements differ. The presented static R,C network analysis of the electrical response has been remarkably successfull when applied to three different cases that have been investigated in some detail.

The static R,C networks representing the electrical response of the ultra pure single crystal silicon and Al-Al₂O₃-Au MIM electron planar emitter are shown in Fig.1 and Fig.2 respectively.

3. Acknowledgements.

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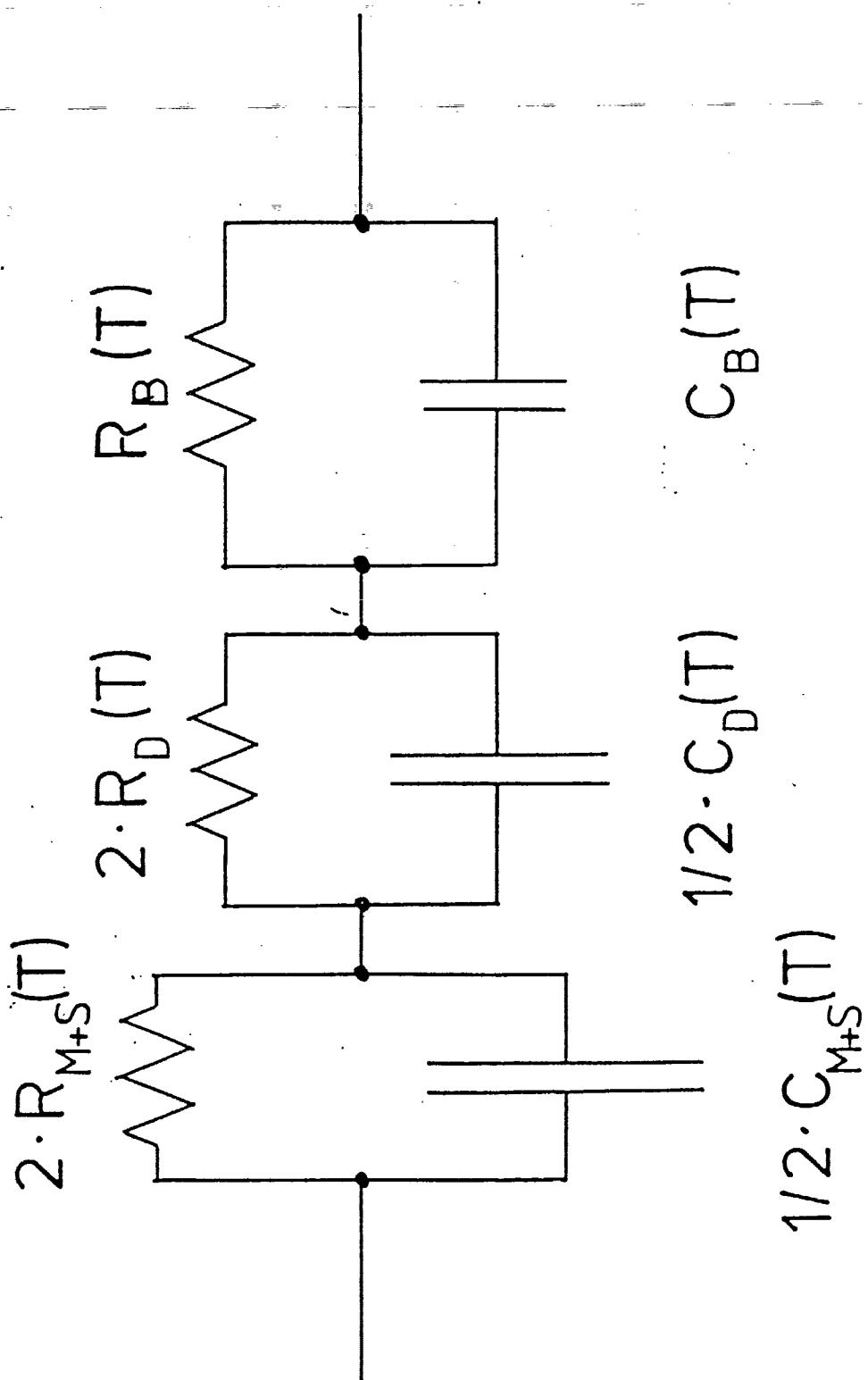
FIGURE CAPTIONS

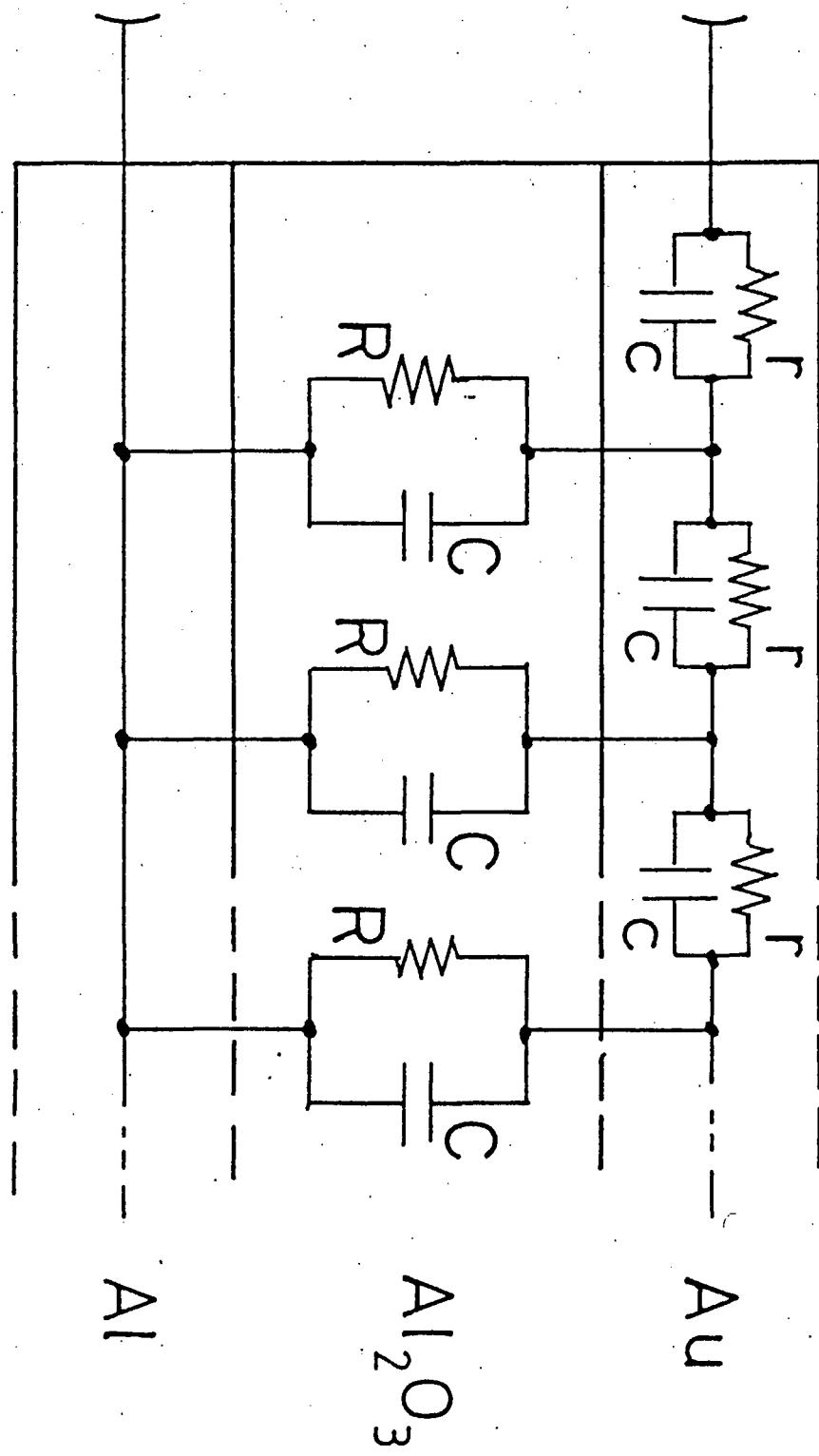
Fig.1. Static R,C network representing the electrical response of ultra pure single crystal silicon.

R_B - electrical resistance of the bulk region
 C_B - geometrical capacitance of the bulk region
 R_D - total electrical resistance of the depletion region
 C_D - geometrical capacitance of the depletion region
 R_{n+s} - quantum mechanical tunnelling "resistance" of the 11.4 Å thick SiO_2 oxide barrier
 C_{n+s} - geometrical capacitance of 11.4 Å thick SiO_2 tunnelling barrier

Fig.2. Static R,C network representing the electrical response of the $\text{Al-Al}_2\text{O}_3\text{-Au}$ MIM planar electron emitter structure

r - electrical resistance of the top thin metal electrode (Au) volume elements
c - geometrical capacitance of the top thin metal electrode volume elements
 C - geometrical capacitance of the Al_2O_3 oxide tunnelling barrier volume elements
R - electrical resistance representing the electrical transport through the Al_2O_3 oxide layer volume elements





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