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# Control System of the Experimental Research of Deposition Process in Vacuum

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### Introduction

Conventional process of thermal evaporation of solids in vacuum involves certain sequence of the technological steps:

initial heating with degasation of evaporator and material(s) in it;

heating up to the desired temperature;

stabilization of the evaporation rate via the temperature control;

exposing the substrate to the vapor (usually shutter is used for this reason)

closing the exposure, when the desirable structure and properties of condensate are reached.

Thickness is one of the essential properties of the deposited thin film. Quartz oscillators and optical interferometer are the most common instruments for in-situ measurements of thin films thickness. However, the surface properties of an oscillator or other measurement probe in most of the cases differ significantly from these, which are at the actual substrate [5]. Therefore above-mentioned thickness measurement methods are limited in sensitivity, especially at the early condensation stages.

One of the common cases for the thickness control of atomic layers deposition is exposure time and vapor pressure control, i.e. open-loop control. The easiest way to control the vapor pressure is to control the temperature of the evaporator. This temperature is influenced by the mass of the material being evaporated and the heat exchange between the evaporator, material and supporting structures [1]. The thermal time constant of the joulic evaporator is comparatively small (20-30 ms), therefore changes in heating current have prompt effect to the temperature of evaporator and, as a consequence, to the vapor pressure.

Obviously, to be accurate in deposition of atomic layers that are thought to be the carrier of the breakthrough potential in today's electronics [2], new methods of thickness control are needed. In this work we disclose the control ideas that were successfully used during our experiments of early stages deposition [3]. They involve the control of an evaporation rate that would be directly related with the vapor pressure, control of the thin film resistance and system for recording the deposition history.

### Control of the evaporation rate

The structure of the evaporation rate (vapor pressure) control system is presented at the Fig. 1. The temperature of evaporator *1* is measured in the indirect way: by measuring the thermo electronic current density with the probes *2* and *3*. The contents of the substance that reaches the probes are the electrically neutral and ionized particles of the evaporated material and the electrons, emitted by both the evaporator and the evaporating material. The density of an electric field at the area of the probes *2* and *3* is dependant on the construction of the evaporator, value and dynamic properties of the current that heats the evaporator, spatial configuration of the grounded parts inside the vacuum camber and the strength and configuration of external electric and magnetic fields. The electromagnetic field is created by the heating current, which ranges from 30 to 300 A. The charged particles that move from the evaporator to the probes are being deflected by this field. Therefore the density of an electric field in the area of the probes *2* and *3* and the substrates *4* has an alternating character and reaches its maximum value, when the heated current crosses the zero value.

The right place for position of probes *2* and *3* in the vacuum chamber is chosen by the experiment, evaluating the actual influence of external electrical and magnetic fields. If probe is close to the evaporator or has large area (it is useful when the evaporation temperature is comparatively low), it has to be cooled or is to have sufficient mass in order not to start emitting the electrons by itself. The measurement of the thermo emission current by the probes *2* and *3* is synchronized with the control of the heating current. The power switch delays the beginning of each half-period of the 50 Hz current, making the time interval, which is sufficient to measure the current density of the thermo electronic emission. Simultaneously the voltages on the substrate and the supplementary probes are measured.

The evaporation rate control system performs two functions: a) degasation of the evaporator and the evaporated material; b) stabilization of the evaporation rate.

At the beginning of the process the temperature of the evaporator is low, and the emission does not start. At this moment the temperature of the evaporator is measured indirectly: by the measurement of the voltage drop over the evaporator $U_g$ and the heating current $i_g$. The value of the average temperature of the evaporator is calculated by the following equation [8]:

$$T(r) = T_0 + \sqrt{\frac{4\alpha_2\left(\dfrac{U_g}{i_g} - r_0\right) + \alpha_1^2 r_0}{4\alpha_2 r_0}} - \frac{\alpha_1}{2\alpha_2}, \quad (1)$$

where $T_0$ – ambient temperature, $U_g$ – voltage drop over the evaporator, $i_g$ – value of the heating current, $r_0$ – value of the "cold" evaporator, measured at the first half period, $\alpha_1$, $\alpha_2$ – temperature coefficients of evaporator material resistivity.

With increase of evaporator temperature $T_g$, gases separate from the heated material. Part of these gas molecules becomes ionized, and is recorded by the probes 2 and 3 as characteristic high-frequency time pattern. When this is sensed by the programmable temperature controller, it temporally switches off the power, until the gases will be pumped out. This is repeated several times, until no gases separation is observed.
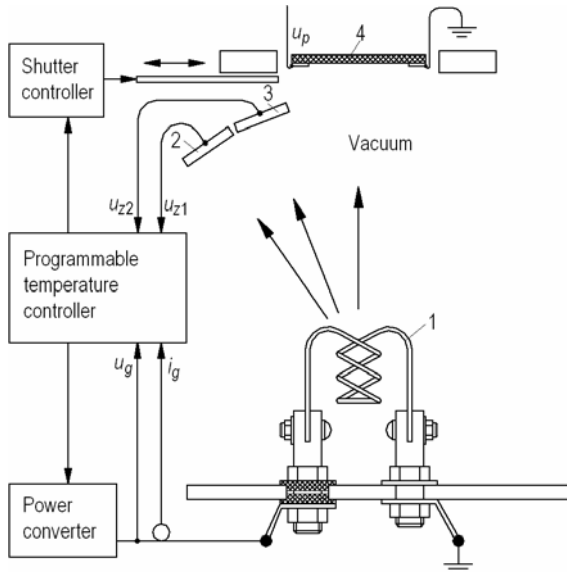


Fig. 1. Structure of the evaporation rate control system

When the temperature of an evaporator reaches the value $T_g$, at which the thermionic emission starts, in the substance that reaches the probes, electrons are dominant. An intensity of these charged particles is dependant on the evaporator temperature and its construction. The surface temperature of the evaporator is calculated from the values $U_{z1}$ and $U_{z2}$ that are measured at the probes 2 and 3 correspondingly with the following equation:

$$T_{gs} = k_k \frac{e(U_{z1} - U_{z2})}{k_B \ln\left(\dfrac{U_{z1}}{U_{z2}}\right)}, \quad (2)$$

where $k_k$ – constructive coefficient, evaluating the spatial position between the evaporator and the probes, $e$ – charge of electron, $k_B$ – Bolcman constant.

This way the temperature $T_{gs}$ is measured when the heating current is zero. After the degasation and initial heating is finished, the temperature controller enables the operation of the shutter controller.

**The system of condensate conductivity control**

Conventional methods of the thin films resistivity measurements employ the well-known four-point network method [4-10], which requires an external power supply. This is rarely acceptable at the initial stages of the condensate formation because of the limited sensitivity: the voltage is limited to the tens of millivolts in order to avoid an influence to the condensate formation properties [9]. The purpose of our conductivity control system is shutting down the exposure, when the desired value of the condensate conductivity is reached. The condensate conductivity is measured by the non-invasive method, which employs the charges, which are present in the vapor stream [1]. The structure of the conductivity control system is presented at Fig. 2. Here the supplementary substrate with two pre-deposited contracts is used as a measurement probe. It can also be the dedicated area at the actual substrate. One of the contacts is connected to the ground, and another is connected to the voltage measurement device, with input resistance $r_m$. We use the instrument with the 10 MΩ input resistance.
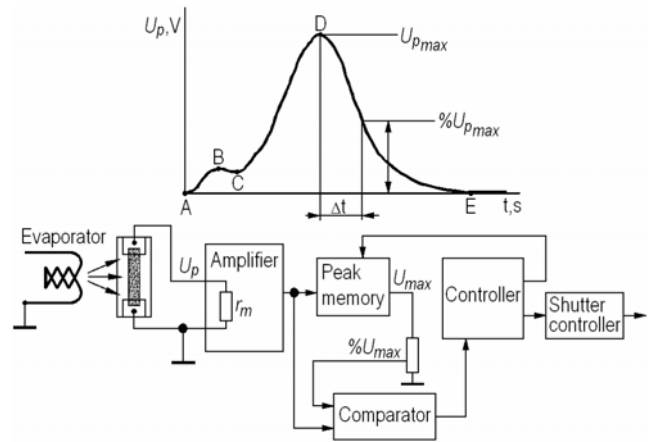


Fig. 2. System of condensate conductivity control

When the shutter controller (Fig. 2) is enabled, it opens the shutter, and the substrate and the measurement probe are exposed to the vapor. It was experimentally determined that when the conductive condensate is being deposited over the substrate, the voltage $U_p$ on the contact a varies with the varying conductivity of the condensate [1,3]. One of the possible time patterns of $U_p$ is shown on the Fig. 2. There are four intervals (they can differ if different surface effects take part): during the interval AB the unstable nucleation is taking the part; at the BC interval the nano-sized clusters of condensate are starting to form. The CD interval can be related to the growing islands of the condensate, which forms continuous thin film at the interval DE.

After our early experimental observations [3], decrease of $U_p$ at the interval DE is unambiguously related with the increase of the condensate conductivity. Therefore the dynamics of $U_p$ at the DE interval was employed for condensate conductivity increase rate measurement and shutter control.

The peak value of the $U_p$ time pattern is detected and remembered by the shutter controller. When the value of the signal starts to decrease, the time measurement is started. The final point for the time measurement is the value of 20-25% from the peak $U_p$. Here two control scenarios are possible: a) if the condensate resistance is sufficient, the shutter is closed; b) if more conductivity is needed or certain amount of conductive monolayers is to be deposited, measured time interval $\Delta t$ is used to calculate the time of the remaining exposure.

**System of recording the history of deposition**

Described surface processes and measurement signals were investigated using the servo-controlled shutter, which enables to "freeze" the deposition stages (Fig. 3). The idea is to partially cover the substrate during the exposure to the vapor in such a way that different areas of the substrate would represent different stages of the condensate formation. This enable the following ex-situ examination of the samples.

The construction of the shutter is shown on the Fig. 4. The stepping motor *2* is mounted on the body *1*, which is firmly attachable to the substrate holder. The helical drive *3* moves the shutter *4*. It is able to cover the substrate *5* in 80 different positions. In parallel to the substrate *5* the measurement probe, made of the same material as a substrate and having two pre-deposited contacts, is fixed. The voltage $U_p$ is measured on the contact *7*, and the contact *8* is connected to the common ground.
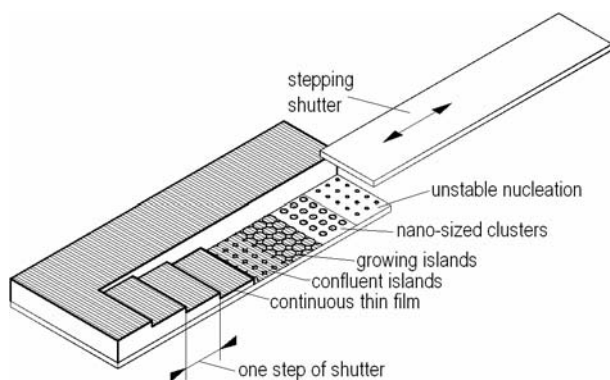


**Fig. 3.** Stepwise coverage of the substrate allow to record the history of deposition

Programmable logic controller with the ability to generate the pulses for shutter movement with the period from 0.02 s was employed to drive the shutter. The pulse rate was set individually for each experiment to match the required linear motion of the shutter.

Minimum step length, achievable by the shutter of described design, is 0.1875 mm. However, due the need to have well-defined areas, representing the history of the deposition, the step length was increased to $1 - 2$ mm. In such a way we avoided the possible diffusion of the vapor

behind the shutter, which leaves 0.1 mm gap to the substrate.

At the beginning of an exposure, when the main shutter is opened, the stepping shutter *4* (Fig. 4) is open, and substrate *5* is exposed to the vapor. Then, in certain time intervals, shutter *4* is being moved over the substrate *5*, covering certain areas of it. The "history" of condensate formation is preserved on the covered area of the substrate.
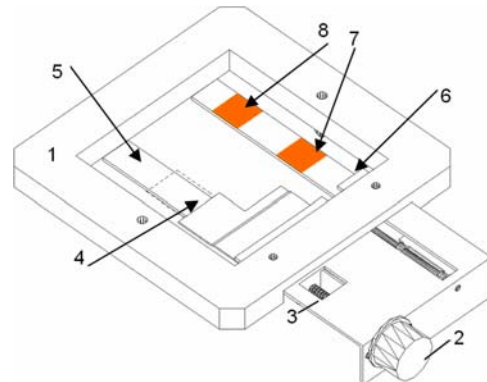


**Fig. 4.** Design of stepping (servo controlled) shutter

We performed several experiments of Cr deposition, using described system of historical record, simultaneously recording the value of $Up$ [3]. The Fig. 5 displays typical time pattern of $Up$ with the time moments, where the shutter was moved to the next position marked. To have an easier navigation during the ex-situ analysis we reduced the number of recorded historical stages to 10, each having 1.875 mm.

Atomic force microscopy (AFM) was used for surface features evaluation. It gave us an opportunity to make the relationship between the $U_p$ time pattern and sample surface features to be evident.
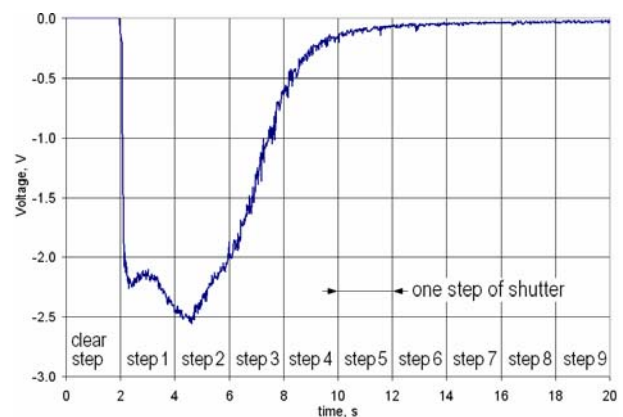


**Fig. 5.** Time pattern of Up value with the shutter movements marked

At the Fig. 6 the sequence of AFM images from the historical stages that are marked in Fig. 5 as *clear step*, *step 2*, *step 3* and *step 4* are shown. After superposition of these images with the results of x-ray photoelectron spectroscopy (XPS) we found the layer-island mode of chromium deposition. This fact slightly conflicts with our schematic model of pure island deposition mode as described earlier. However, informativity of $U_p$ time pattern is without any

doubts evident and was successfully related to the conductive properties of deposited thin film [3].
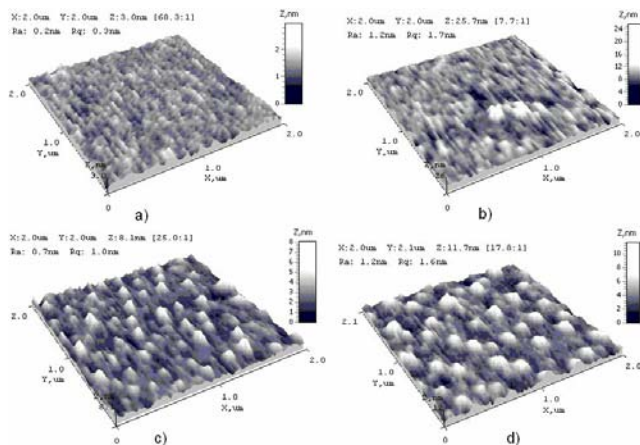


**Fig. 6.** AFM (NT-206) images of: a and b – early nucleation stages; c – stage of coalescence of islands; d – continuous film

### Conclusions

1. Proposed system of thin films properties control, consisting of vapour pressure control, film conductivity control and stepwise coverage control subsystems enables the production of thin films with near atomic thickness with highly repeatable properties.

2. The servo controlled shutter allows to "freeze" the historical stages of thin film deposition with high reliability without interrupting the deposition process.

### References

1. **Virzonis D., Sinkevicius V.** Increasing the observability of the thin films manufacturing process // Electronics and Electrical Engineering. – 2004. – No. 4(53). – P. 42–45.
2. **ENIAC** strategic research agenda. 2005, European Council.
3. **Jukna T., Baltrušaitis J., Sinkevičius V., Viržonis D.** Verification of early condensate formation stages monitoring: the case study // Submitted to Thin Solid Films; to appear in 2007.
4. **Brajpuriyaa R., Tripathi S., Sharma A., Shripathi T., Chaudhari S. M.** Thickness dependent structural and magnetic properties of ultra-thin Fe/Al structures // The European Physical Journal B. – 2006. – No. 51. – P. 131–136.
5. **Dobierzewska-Mozrzymas E., Pieciul E., Bieganski P., Szymczak G.** Conduction Mechanisms in Discontinuous Pt Films // Crystal Research Technology. – 2001. – No. 8–10(36). – P. 1137–1144.
6. **Burkov A. T., Vinzelberg H., Schumann J., Nakama T., Yagasaki K.** Strongly nonlinear electronic transport in Cr-Si composite films // Journal Of Applied Physics. – 2004. – No. 12(95). – P. 7903–7907.
7. **Bishay A. G., Fikry W., Hunter H., Ragie H. F.** Temperature coefficient of the surface resistivity of two-dimensional island gold films // J. Phys. D: Appl. Phys. – 2000. – V.33. – P. 2218–2222.
8. **Dayal D., Rudolf P., Wibmann P.** Thickness dependence of the electrical resistivity of epitaxially grown silver films // Thin Solid Films. – 1981. – V.79. – P. 193–199.
9. **Schuisky M., Elam J. W., Georgea S. M.** In situ resistivity measurements during the atomic layer deposition of ZnO and W thin films // Applied Physics Letters. – 2002. – No. 1(81). – P. 180–182.
10. **Palasantzas G., Zhao Y., Wang G., Lu T., Barnas J., De Hosson J. Th. M.** Electrical conductivity and thin-film growth dynamics // Physical Review B. – 2000. – No. 16(61). – P. 109–117.

**V. Sinkevičius, D. Viržonis, L. Šumskienė, T. Jukna. Control System of the Experimental Research of Deposition Process in Vacuum // Electronics and Electrical Engineering. – Kaunas: Technologija, 2007. – No. 5(77). – P. 7–10.**

The control system for near-atomic thin films deposition is disclosed. The system is consisting of three parts: vapor pressure control, film conductivity control and stepwise coverage control. All three subsystems are explored experimentally and found to have the sufficient potential to give improved repeatability of deposition of near-atomic thin films. The results and conclusions from stepwise coverage experiments, followed by AFM and XPS analysis is presented as an evidence of direct relationship between the in-situ recorded voltage pattern and the film formation stages. Ill. 6, bibl. 10 (in English; abstracts in English, Russian and Lithuanian).

**В. Синкявичюс, Д. Виржонис, Л. Шумскене, Т. Юкна. Система управления экспериментального исследования процессов конденсации в вакууме // Электроника и электротехника. – Каунас: Технология, 2006. – № 5(77). – С. 7–10.**

Раскрыта система управления процессом конденсации плёнок толщиной от нескольких до десятков атомов. Она состоит из трёх частей: система управления давления паров, система измерения проводимости конденсата и система пошагового покрытия. Все три системы исследованы экспериментально и установлено, что они способны увеличить повторяемость плёнок толщиной в несколько атомов. Результаты пошагового покрытия исследовались на AFM и XPS и было установлено, что существует связь между измеренным сигналом и состоянием конденсата. Ил. 6, библ. 10 (на английском языке; рефераты на английском, русском и литовском яз.).

**V. Sinkevičius, D. Viržonis, L. Šumskienė, T. Jukna. Vakuume vykstančių kondensacijos procesų eksperimentinio tyrimo valdymo sistema // Elektronika ir elektrotechnika. – Kaunas: Technologija, 2007. – Nr. 5(77). – P. 7–10.**

Atskleista plonų sluoksnių, turinčių keleto ar kelių dešimčių atomų storį, nusodinimo valdymo sistema. Ją sudaro trys dalys: garų slėgio valdymo sistema, sluoksnio laidumo sistema ir žingsninio dengimo sistema. Visos trys sistemos ištirtos eksperimentiškai ir nustatyta, kad jos turi pakankamą potencialą padidinti atominį storį turinčių kondensuotų plėvelių savybių pasikartojimą. Žingsninio dengimo eksperimentai, kuriuos lydėjo AFM bei XPS analizė, parodė, jog teiginys, kad egzistuoja tiesioginis ryšys tarp kondensacijos proceso metu užregistruotos įtampos ir plėvelės formavimosi etapų, yra pakankamai pagrįstas. Il. 6, bibl. 10 (in English; summaries in English, Russian and Lithuanian).