

LOW-COHERENT LIGHT INTERFEROMETRY FOR MONITORING OF TECHNOLOGICAL PROCESSES IN A METALORGANIC VAPOR-PHASE EPITAXY

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Abilities of low-coherent tandem-type interferometry in optical monitoring of the substrate temperature and bending during metalorganic vapor-phase epitaxy (MOVPE) were demonstrated for the first time. Thickness of growing layer was monitored as well. Absolute precision in measuring the temperature of the Si, GaAs or sapphire substrates during the MOVPE process is limited by the calibration accuracy and reaches ± 1 °C. The error of *in situ* estimation of the layer thickness is 2 nm. Dramatic disagreements (in the range of 10-100 °C depending on pressure, gas consumption, temperature etc) between readings of the thermocouple fixed inside the graphite substrate holder and the actual substrate temperature were revealed. Furthermore, we detected significant temperature gradients (along the substrate holder) that depend on the carrier gas consumption and other factors.

1. Introduction

Modern technologies for manufacturing of micro- and nanostructures and devices display an increasing importance of reproducibility of processes; efforts are being made to improve the product yield in a single process. This demands the necessary presence of systems for *in situ* control of the technologically important parameters [1]. For the most processes (including film depositing, annealing and etching of the obtained thin film structures), fundamental parameters are the substrate temperature and the rate of growth or etching. At present, the substrate temperature is routinely controlled by thermocouples or pyrometers. However, the accuracy of these measurements is fairly low. In the first case, it is because of the absence of a good, reproducible thermal contact, especially in vacuum. In the second case, it is associated with uncertainty of the source of incident irradiation and with the variation in the optical properties of the objects themselves which may markedly vary during the entire process. Alternatively, the growth temperature can be traced from the dependence of the semiconductor bandgap on the temperature [2]. However, temperature of a multilayer compound structure is hardly measured precisely by using this method.

First attempts to utilize optical methods for *in situ* control of the epitaxial growth of semiconductors were undertaken almost 20 years ago [3,4]. Nowadays, changes of surface anisotropy under the epitaxial growth of layers [5,6], and changes in the reflection coefficient which result from the probing light interference inside the substrate [7-9] or the growing film [10] are optically registered. In

the latter case, a broadband light source with the subsequent spectral analysis is often applied [11]. Nonetheless, an accurate determination of both the temperature and the rate of semiconductor epitaxial growth in the processes of molecular-beam epitaxy (MBE) or MOVPE represents so far a very complicated problem, keeping in mind that these parameters should be monitored simultaneously.

In our opinion, a new type of optical instrumentation [12,13] is very promising. It uses the principle of low-coherent tandem interferometry at which the probing light successively goes through the sample under investigation (first interferometer) and an optical correlometer (second interferometer). The difference in optical path can be tuned with a very high precision. Advantages of such a principle are the following: high and continuous sensitivity in all the range of the measured optical thicknesses including the initial stage of film growth; possibility of carrying out absolute measurements; and the universality since it enables to measure the thickness and the temperature on practically any kind of substrates. This work is aimed at adaptation and utilization of this type of instrumentation for *in situ* control of the temperature and the thickness of growing structures in a MOVPE reactor using GaAs, Si and sapphire substrates.

2. Experimental part

2.1. The operating principle of a low-coherent tandem interferometer

The schematic of a low-coherent tandem interferometer is shown in Fig.1. The equipment consists of a low-coherent source of light and two interferometers (delay lines) optically coupled through an optical fiber. If differences in the optical path-length of both interferometers are larger than the length of coherence of the light source, then neither of them taken separately produces an interference signal. However, the interference will occur if differences of optical path-length L_1 and L_2 coincide as a result of low-coherent light transmission through both delay lines.

This is realized by the following way. The first interferometer divides the coherence pulse into two non-overlapping pulses with the delay L_1 between them. At the input of the second interferometer each of these pulses is divided into another two pulses. As a result, we have four pulses at the output of the second interferometer. On the assumption of

$$\Delta L = |L_2 - L_1| < L_{coh}$$

where L_{coh} – the length of a source coherence, two of the four pulses will overlap and interfere.

Therefore, adjusting the length of one of the delay lines (reference) to the maximum of the interference picture at the output of the device, one can fix the optical thickness of the other delay line (measured object). This allows monitoring of its changes during the growth or etching of the sample, or the temperature changes. At that, absolute accuracy of the optical thickness measurement is limited by the accuracy of the device used to register the path difference occurring in the reference delay line, whereas the sensitivity to small changes is limited by its stability.

The given principle has been used by us to design the instrumentation for an efficient technological control of the float-glass thickness at hot stages of its production [14,15]. By now, several models of the developed system have been installed and successfully exploited at factories in Russia and “near abroad”.

2.2. Calibration

The first problem which has to be solved when measuring the absolute temperature of substrate is to obtain the calibration dependence of the substrate optical thickness (D) on the temperature in the operating temperature range:

$$D(T) = n(T)d(T) = D_0(1 + f(T)),$$

where n – index of refraction, d – geometric thickness, D_0 – optical thickness at some initial temperature T_0 .

It should be noted here that in low-coherent circuits two characteristics of an interference signal change with varying temperature: position of the maximum of envelope and the filling phase. The change of the first characteristic is caused by the change of the group refraction index n_{g1} , whilst the second – by phase refraction index n_{ph} . Dispersion of the refraction index leads to the difference between dependences $n_g(T)$ and $n_{ph}(T)$. To control small changes in the optical thickness the measurements of phase are preferable since they are more accurate. However, at large temperature changes the phase measurements become less convenient. This is because the phase is determined as accurate as $2\pi N$, where N – number of the interference fringe, so the information will be lost during the uncontrolled jump from one fringe to another. Besides, the long-time stability of phase of the probing light might appear insufficient since the phase depends on the wavelength of superluminescence diode (SLD) and changes with time under influence of various external factors (temperature, pumping current, ageing of SLD itself etc). That is why the calibration is to be done for the group refraction index namely.

To determine the calibration dependence $f(T) = (D - D_0)/D_0$ the substrate was placed inside the thermostat, which was a massive copper furnace with resistive heating. The furnace temperature was controlled by a thermocouple fixed in the inside and was kept constant using a feedback system with accuracy of $\pm 1K$. Input-output of the probing radiation was realized through a small quartz window. The diameter of the probing light spot on the substrate was less than $50 \mu m$. The experimental dependence $f(T)$ (Fig.2) was obtained for the following substrates: $379 \mu m$ thick silicon wafers KDB-15; $388 \mu m$ thick silicon wafers KEF-4.5; a semi-insulating GaAs substrate with the thickness of $443 \mu m$; and a sapphire substrate polished at both sides with the thickness of $422 \mu m$ which was cut out perpendicularly to the optical axis (c -orientation).

The experimental data show that

- a) doping of the silicon wafer does not affect the temperature dependence of the optical thickness in the temperature range of 300-800K (within $\pm 1K$ interval which is defined by the equipment for temperature maintenance);
- b) with the accuracy given above, the temperature dependence of the optical thickness for silicon in the temperature range of 300–800K ($\lambda=1550nm$) is described by formulae:

$$f_{Si}(T) = 3.486 \cdot 10^{-8} T^2 + 4.455 \cdot 10^{-5} T - 1.65 \cdot 10^{-2}, T_0 = 300 K;$$

- c) the temperature dependence of the optical thickness for GaAs in the temperature range of 300-600 K@1310nm is described by formulae:

$$f_{GaAs}(T) = 6.369 \cdot 10^{-8} T^2 + 7.407 \cdot 10^{-5} T - 2.795 \cdot 10^{-2}, T_0 = 300 K;$$

d) the temperature dependence of the optical thickness for sapphire in the temperature range of 400–1400 K at the wavelength of 1550nm is described by formulae:

$$f(T) = 4.08 \cdot 10^{-9} T^2 + 1.178 \cdot 10^{-5} T - 3.903 \cdot 10^{-3}, T_0 = 300 \text{ K}.$$

As the measured value is an average of the temperature throughout the substrate, the question about the difference of temperatures between the front (operating) and the rear surfaces arises. The difference of temperatures is easy to estimate considering that the density of the heat flux going through the substrate and graphite holder is constant and neglecting the differences between radiative abilities of graphite and substrate.

Starting from heat conduction equation

$$\mathbf{h} = -c \nabla T,$$

where $\mathbf{h}$ – heat flux density, c - heat conduction, the temperature gradient for a substrate is possible to express as

$$DT = \mathbf{h}/c \cdot d.$$

Here d is substrate thickness. Taking the power absorbed by the substrate holder at the operating temperature $\sim 680^\circ\text{C}$ equal to $\sim 1 \text{ kW}$, the substrate thickness equal to $\sim 400 \mu\text{m}$, and $c_{\text{Si}} = 30 \text{ W/(mK)}$ at the temperature of 950 K [16], we obtain the estimation of maximal DT as 1.3 K.

2.3. Experiments in a horizontal reactor.

Studies of technological parameters in the horizontal-type reactor were conducted with an Epiquip plant, model VP502-RP (the diagram of experimental setup is given in Fig.3) equipped with induction heating of the graphite substrate holder. The measuring head was located at the height of 30cm from the substrate. Optical measurements were taken from three points at the beginning, middle and end of the substrate holder.

Fig.4 shows the dependences of the temperature differences for substrates located in different parts of the substrate holder and readings of the thermocouple $\Delta T = T_{\text{nod}} - T_{\text{m/n}}$ at different temperature in vacuum and in hydrogen flux (3 l/min) at the pressure of 100 mBar. In vacuum, readings of the thermocouple fixed inside the substrate holder (this thermocouple was included in the feedback system for automatic maintaining the temperature of holder) proved to be very overrated in comparison with the real temperature of the substrate. This should evidently be assigned to the lack of a good thermal contact between the holder and the substrate. The presence of the hydrogen carrier gas makes for a the better thermal contact and accordingly reduces ΔT .

As follows from our data, the central part of the substrate holder is the hottest area. This is obviously connected with the less strength of electromagnetic field created by the inductor near edges and with additional losses due to heat release from the faces of the substrate holder. At the same time, the temperature gradient increases with increasing temperature. The observed “overheating” of the center of substrate holder in presence of hydrogen is attributed to the fact that the thermocouple is located in the low temperature region in the beginning of the substrate holder.

The increase of hydrogen flux from 3 l/min to 8 l/min causes the alterations in temperature distribution along the substrate holder (Fig.5). The substrate located in the beginning of the holder becomes 16 degrees colder due to the additional cooling by the oncoming flux of the cold carrier gas. Meanwhile, the temperature of the substrate located at the end of the holder increases by 2 degrees. Note that the feedback system keeps the temperature of substrate holder virtually constant, as measured at the thermocouple junction (Fig.6). Switching of gas fluxes is indicated by arrows in Fig.6. The initial section of the graph reflects the process of establishing the operating temperature of 683 °C under hydrogen flux of 3 l/min.

It is interesting to compare the time required for establishing the operating temperature for different types of substrates, especially when the thermal contact between the heater and the substrate is not good enough, for example, in vacuum (*i.e.*, the substrates are heated due to the absorption of thermal radiation). We investigated the process of heating silicon and sapphire substrates to the temperature of 300 °C in vacuum (10^{-5} mBar) and to the temperature of 700 °C at the reduced pressure of 100 mBar. Since sapphire substrates are transparent, their rear side is usually covered with a molybdenum film in order to increase the absorption coefficient in the IR domain and hence reduce the time required to reach the operating temperature. As our measurements showed, in vacuum the heating time required for establishing the temperature of 300 °C for sapphire substrates was shorter than that for silicon substrates. Most likely, this can be explained by the presence of a strong absorption band of sapphire in the range of 6 μm . At the pressure of 100 mBar, the heat transfer occurs through the carrier gas, so that the time required for establishing the substrate temperatures is the same in both cases, up to the temperature of ~ 700 °C.

Fig.7a shows the changes of the optical thickness of epitaxial structure on the GaAs substrate at first stages of growth. The decrease of the optical thickness after interrupting the growth is due to decrease of the substrate temperature by about 0.3°C after mixing the hydrogen flux with silane (0.5 l/min) in order to form a doped δ -layer. The instrumentation sensitivity (mean square deviation from average) to the changes in the growing film thickness is 2 nm, the rate of growth – 54 nm/min (in particular process). Fig.7b presents the changes of the optical thickness of the GaAs substrate during the epitaxial growth of the GaAs/AlAs heterostructure. In this process, the instrumentation sensitivity (mean square deviation from average) to the increase in the film thickness was equal to 10 nm, the rate of GaAs growth – 13.5 ± 2 nm/min, and the rate of AlAs growth – 37 ± 2 nm/min.

2.4.Experiments in a vertical reactor.

Technological parameters in the vertical-type reactor were studied using a self-made apparatus designed for the growth of GaN based wideband materials (diagram of the experimental setup is given in Fig.8). Here, the molybdenum substrate holder was heated inductively. The measuring head was mounted at the height of 30 cm from the substrate. The input-output of the probing radiation was implemented through the quartz optical window OW located in the upper part of the reactor (Fig.8).

Fig.9 shows the signal recording (finger-print) during growth process of the GaN film beginning from heating and annealing the sapphire substrate at the tem-

perature of 1100 °C (stage #1). Then follows the stage of nitridization at the temperature of 550 °C (stage #2); stage #3 of the growth of buffer GaN layer with the thickness of 100 nm at temperature 550 °C (the inset shows the changes of thickness “in phase”); stage #4: heating the substrate up to 1050 °C; stage #5: growth of GaN layer; and stage #6 completes the process with cooling. These stages are separated by vertical lines in Fig.9.

The 13th minute of the process (stage #1) shows the decrease of the temperature by 12 °C which is associated with the substrate cooling due to ammonia feeding (0.2 l/min). At the 24th minute, the ammonia consumption was increased up to 2 l/min which led to the appearance of nonstationary vapor flux in the reactor and in the rise of a noise while taking measurements of optical thickness from the substrate “along the envelope”. It should be noted that in absence of the splitter *Sp* (Fig.8) the nonstationarity occurs at significantly lower ammonia consumption, beginning from 0.4-0.5 l/min. The 40th minute, after completing the buffer layer growth, was followed by heating up to the operating temperature of 1050 °C. This was accompanied by bending of the substrate upwards. Fig.9 demonstrates abrupt increase of the gap size between the lower surface of the substrate center and the substrate holder by almost 400 µm (the substrate diameter is 2 inches). The reasons of such a behavior are not yet clearly understood and, as it was mentioned earlier [17], require additional studies. The differences in the object optical thickness (substrate + film) measured “along the phase” and “along the envelope” become more pronounced with increasing thickness of the growing GaN film. The optical thickness “along the phase” changes practically linearly with time whereas the optical thickness “along the envelope” oscillates. These oscillations are connected with an interference of the probing light in inside of the growing film and result in distorting the form of the received signal envelope, and namely in shifting the maximum of this envelope. The amplitude of oscillations increases as the surface morphology improves, due to equalizing the phase of the probing light received from different sections of the illuminated spot on the substrate. From the number of oscillation maximums (minimums) and taking into account the refraction index of GaN ($n=2.4$ at the wavelength 1.55 µm) we obtain the thickness of grown film equal to 2.68 µm, that is in reasonable agreement with the measurements “along the phase” (2.71 µm).

3. Conclusion

By using new principles of registering the optical thickness of transparent wafers we introduced new high-accuracy instrumentation for *in situ* optical control of the real substrate temperature and bending, as well as the thickness of growing layers during the epitaxial growth of heterostructures by MOVPE or in vacuum. Substrates made from GaAs, Si, or sapphire were utilized in this study. Wide disagreement (10-100°C) between the readings of the thermocouple (mounted inside the substrate holder) and the temperature measured by the optical method was observed. The disagreement rises with increasing temperature of the substrate and flux of the carrier gas, as well as with reducing pressure in the reactor. In the horizontal-type reactor, significant gradients of the temperature along the substrate holder were detected.

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Fig.1 Schematic of a low-coherent tandem interferometer. SLD – a low-coherent source, F – a connecting optical fiber, PD – photodetector, $L_{1,2}$ – an optical path difference of the first and the second interferometers, accordingly, BS – light dividers, M – mirrors.

Fig.2 The dependence of increase in optical thickness on temperature $f(T)$ for different materials. Dots – experimental data, lines - approximation.

Fig.3 The diagram of the experimental setup in a horizontal reactor. M – measuring unit, PC – computer, F – optical fiber, I – induction coil, Obj – objective, S – the measured substrate, Sus – a substrate holder.

Fig.4 The difference between the substrate temperature and thermocouple readings depending on the temperature measured by the thermocouple.

Fig.5 The difference between the substrate temperature and thermocouple readings depending on the position the observation point along the substrate holder at $T=680^{\circ}\text{C}$.

Fig.6 The dependence of temperature on time demonstrating changes for the substrate located in the beginning of the holder with varying hydrogen consumption.

Fig.7 Changes in optical thickness of the GaAs substrate in the MOVPE process: a) epitaxy of GaAs based geterostructure for Shottky diode; b) epitaxy of a GaAs/AlAs heterostructure.

Fig.8 The diagram of the experimental setup in a vertical-type reactor. M – measuring unit, PC – computer, F – optical fiber, I – induction coil, Obj – objective, S – the measured substrate, Sus – a substrate holder, OW – optical window, Sp – splitter.

Fig.9 Finger-print of the technological process for the MOVPE growth of GaN film on sapphire substrate. a) —○— optical thickness according to the measurements “along the phase”, —△— optical thickness according to the measurements “along the envelope”, b) substrate bending.

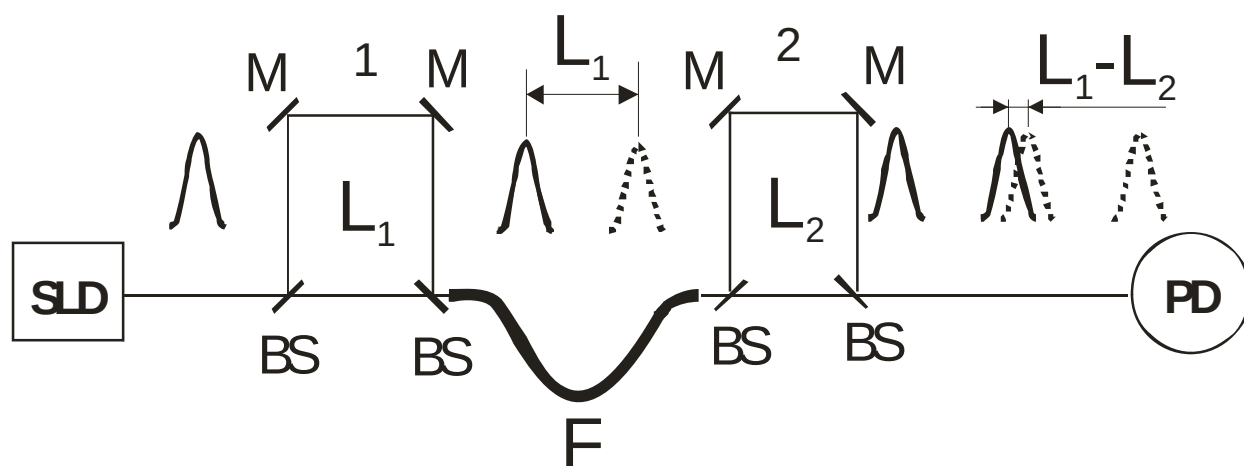


Fig.1.

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Optical monitoring of technological process parameters under the conditions
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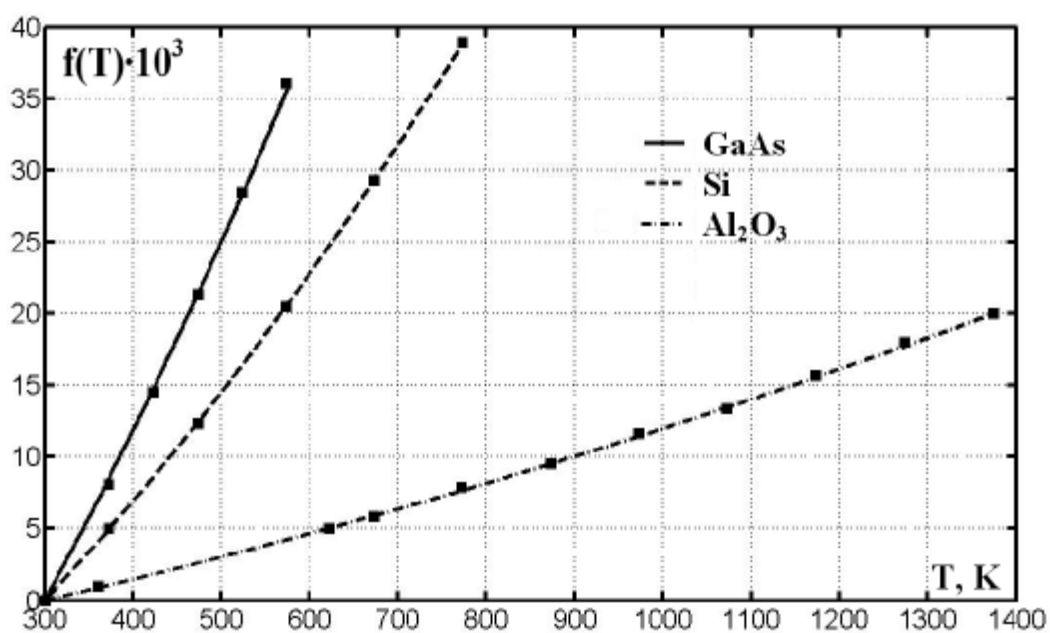


Fig.2.

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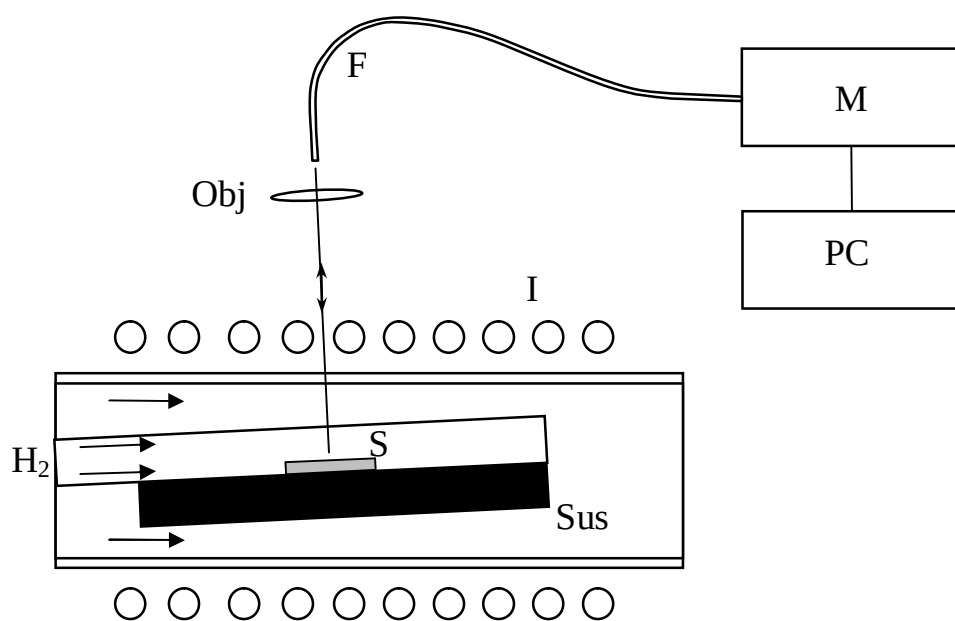


Fig.3.

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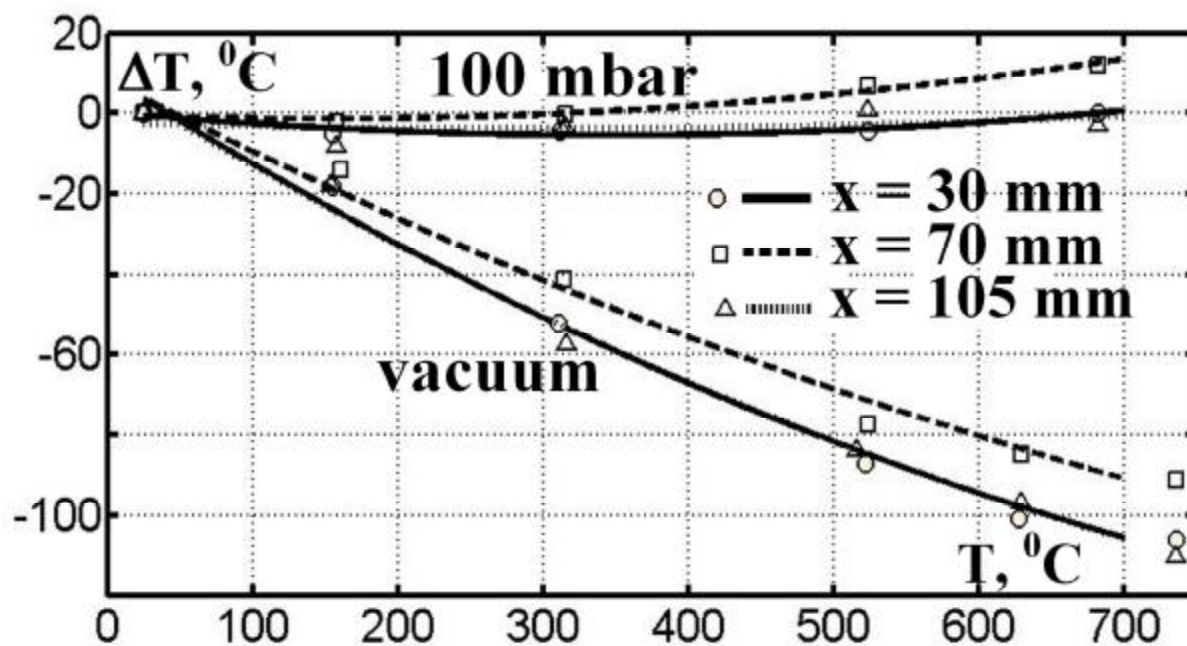


Fig.4.

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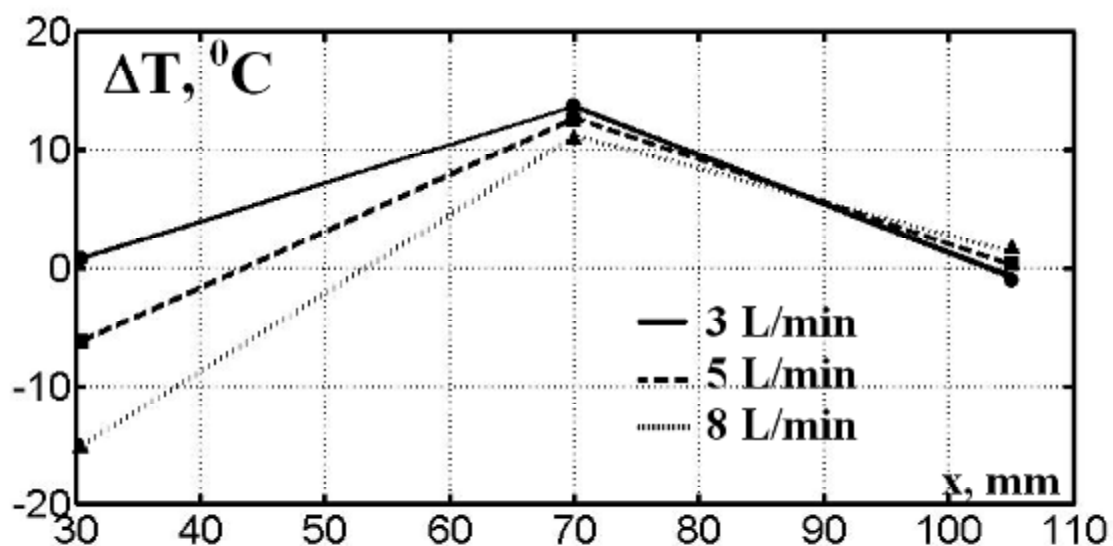


Fig.5.

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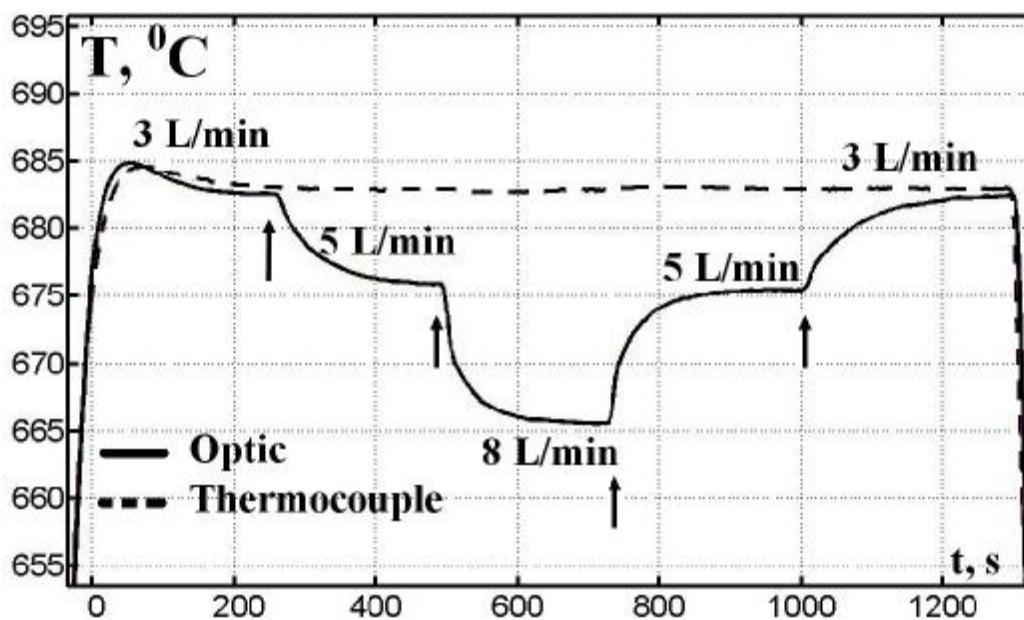
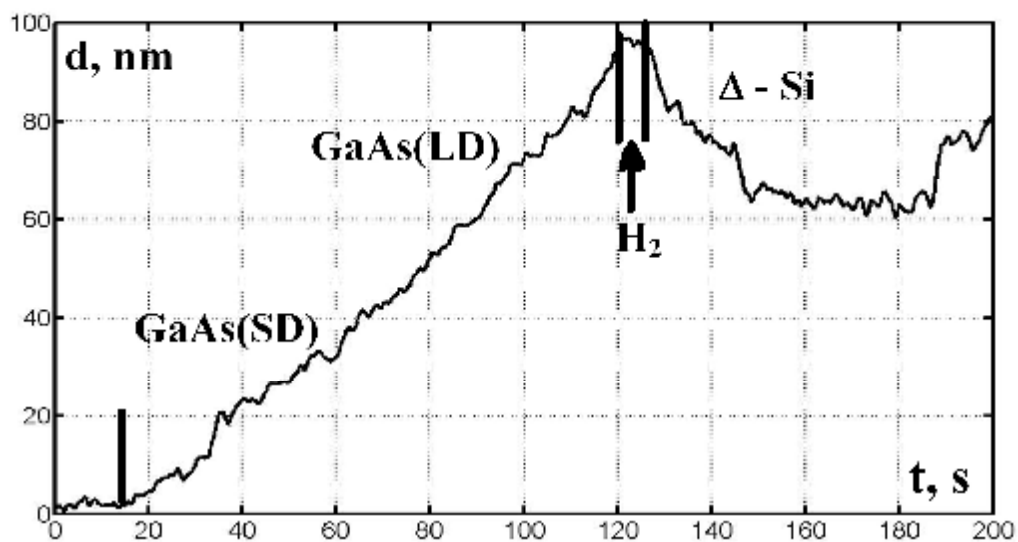


Fig.6.

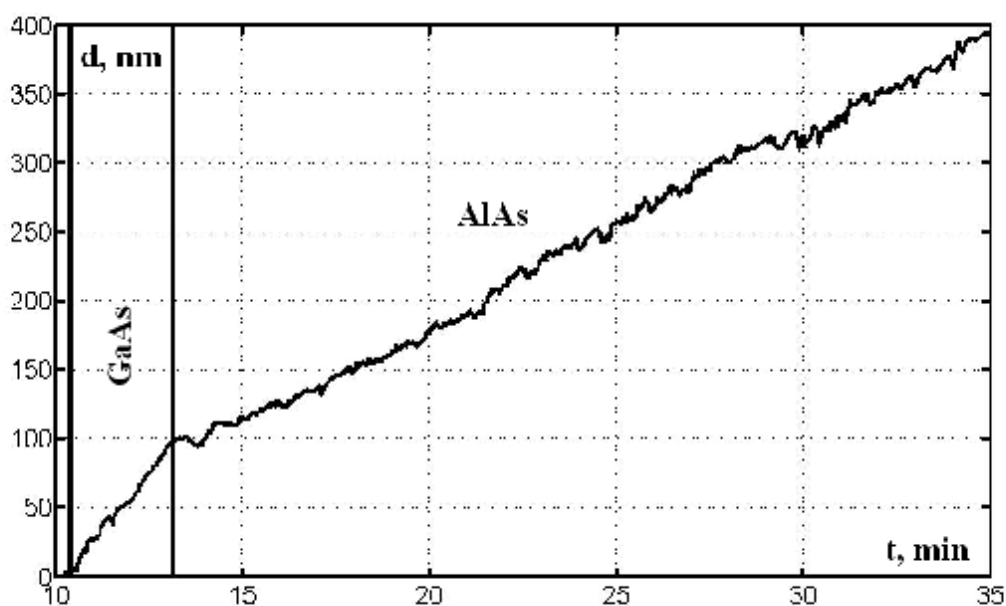
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a)



b)

Fig.7.

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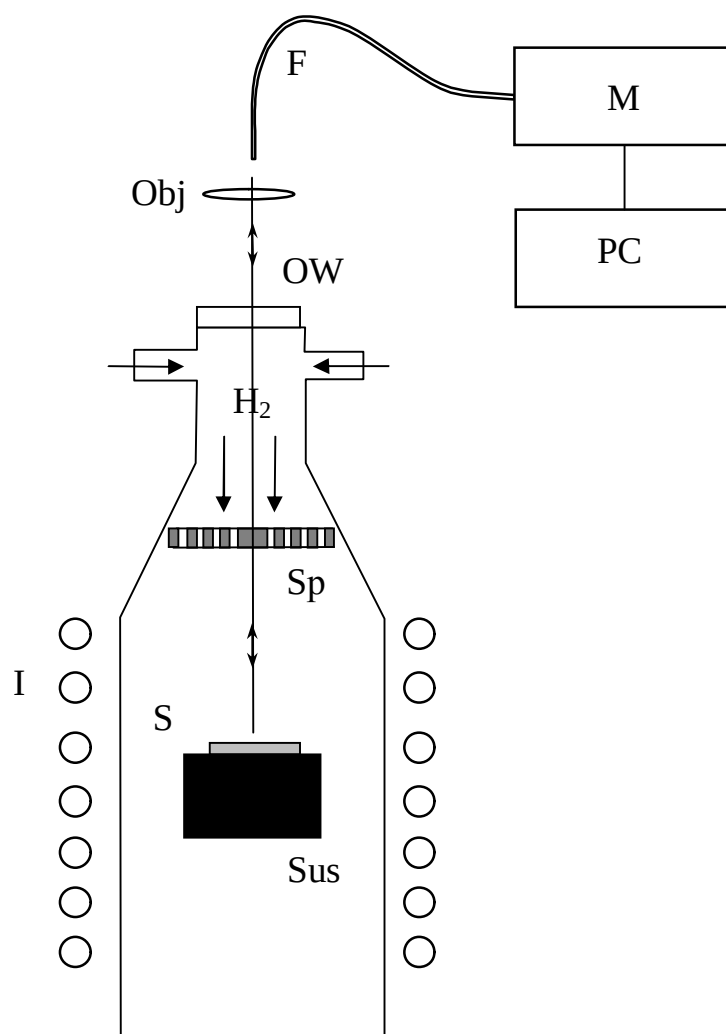


Fig.8.

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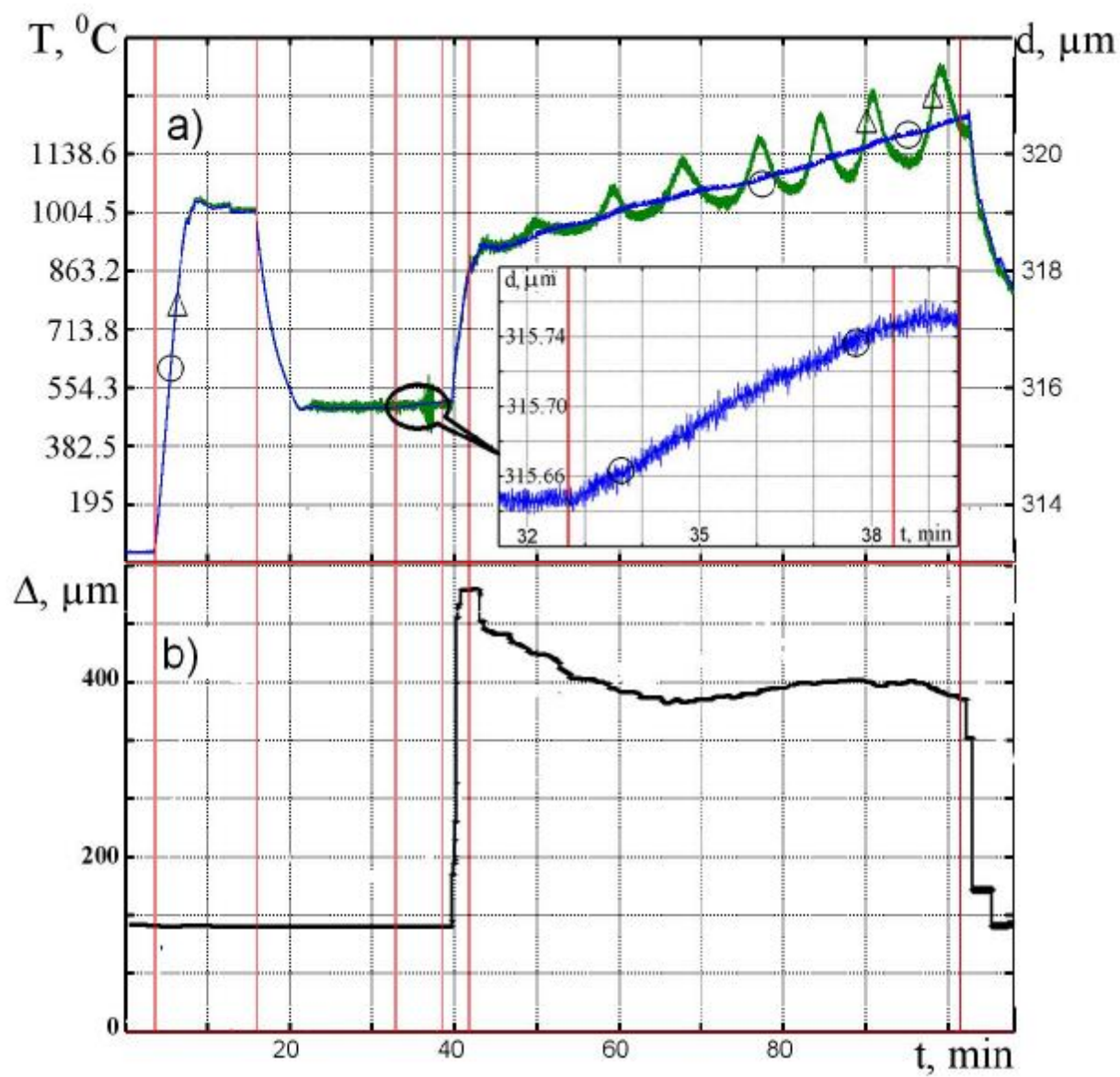


Fig.9.

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