ISSN 0020-4412, Instruments and Experimental Techniques, 2017, Vol. 60, No. 5, pp. 759–763. © Pleiades Publishing, Ltd., 2017.
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LABORATORY TECHNIQUES

An Ultrahigh-Vacuum Multifunctional Apparatus for Synthesis and in Situ Investigation of Low-Dimensional Structures by Spectral Magnetoellipsometry in the Temperature Range of 85–900 K

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Received October 10, 2016; in final form, January 27, 2017

Abstract—This paper presents the results of modernizing an ultrahigh-vacuum multifunctional apparatus that allows one to obtain semiconductor or metallic nanostructures in a single technological cycle and to investigate their optical and magneto-optical properties in a temperature range of 85–900 K. The capabilities of the developed system were demonstrated based on the example of studying the temperature dependence of the bulk Si permittivity via spectral ellipsometric measurements.

DOI: 10.1134/S0020441217050086

The continuous development of the microelectronic industry towards miniaturization, energy saving, and an increase in the working capacity of electronic devices provides a stable demand for new materials, their synthesis technology, their properties.. As an example, the application of the methods of molecular-beam epitaxy (MBE) of semiconductors makes it possible to produce high-quality structures of field-effect transistors for electronic devices [1], while the epitaxy of ferromagnetic materials is a method to design devices of spintronics [2]. In addition to the synthesis methods, methods for in situ analysis of the structural, optical, and magnetic properties of created materials are important as well. Non-destructive in situ analytical methods are especially important, because investigations of synthesized nanostructures require an ultrahigh vacuum that prevents the sample contamination and oxidation processes.

In the recent decades, the ellipsometry method has proved itself in the role of an efficient non-destructive contactless analytical method [3]. The use of spectral ellipsometry in the range of 1.5–6 eV with application of an external magnetic field to a sample actually combines the ellipsometric and magneto-optical analytical methods, thus allowing one to determine both the optical properties and the surface morphology [4], and investigate the electron structure and magnetic properties of synthesized ferromagnetic materials [5]. However, the in situ methods of the analysis with the possibility of controlling and specifying the temperature of a sample in a wide range from units of Kelvin to heat-induced sample destruction are of greatest research interest. As an example, in the case of ferromagnets, analyzing the temperature dependences of the saturation field and the coercive force from magneto-optical measurements makes it possible to determine the phase composition of a ferromagnetic structure and the behaviour and type of domain boundaries [6].

The analysis of the literature data shows that the in situ monitoring of the temperature dependences of the optical properties allows: (1) the observation of solid-phase reactions and changes in the sample-surface morphology [7]; (2) the control of the annealing technology and the progress of interlayer reactions in various nanostructures by measuring the temperature dependences of the spectrograms of the diagonal components of the permittivity tensor; (3) the analysis of the spin-polarized density of electron states in a ferromagnet by measuring the spectral dependences of nondiagonal components of the permittivity tensor at low temperatures [5]; (4) determination of the critical points of the Brillouin zone in semiconductor silicides in investigations of the electron–phonon interaction [8]; and (5) the acquisition of information on the influence of the film–substrate interface on the spin polarization of conduction electrons via recording of the temperature-dependent amplitudes of the absorption bands of the magnetic circular dichroism [9].

This study was aimed at the development and tests of an ultrahigh-vacuum multifunctional apparatus that makes it possible to synthesize and study low-dimensional structures in situ using the spectral mag-

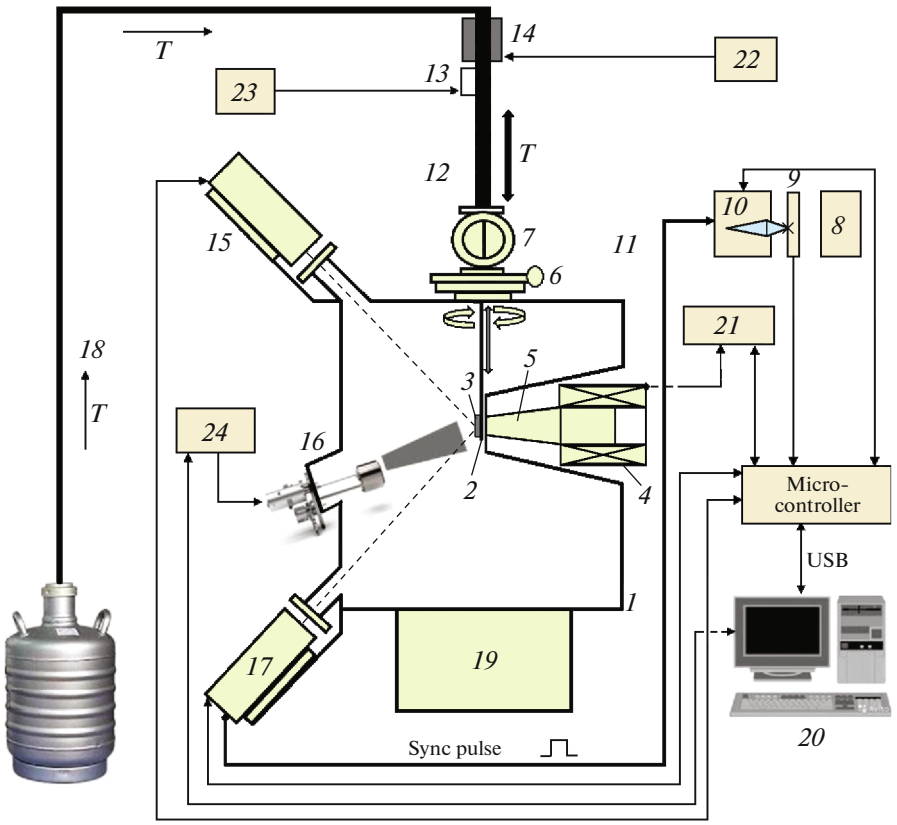

Fig. 1. A block diagram of the ultrahigh-vacuum multifunctional apparatus: (*1*) vacuum chamber; (*2*) rod with the sample holder; (*3*) investigated sample; (*4*) electromagnet; (*5*) magnetic circuit; (*6*) slide gate; (*7*) lock chamber for sample recharging; (*8*) light source; (*9*) light chopper; (*10*) monochromator; (*11*) light guide; (*12*) linear bellows translator; (*13*) vacuum bellows three-degree-of-freedom manipulator; (*14*) flow-type evaporator–heater; (*15*) analyzer unit; (*16*) molecular source; (*17*) polarizer unit; (*18*) nitrogen supply system; (*19*) magnetic-discharge pump; (*20*) computer; (*21*) magnet power-supply unit; (*22*) flow-type heater power-supply unit; (*23*) sample-heater power-supply unit; and (*24*) evaporator power-supply unit.

netic-ellipsometry method in the temperature range of 85–900 K.

THE ULTRAHIGH-VACUUM MULTIFUNCTIONAL APPARATUS

The ultrahigh-vacuum multifunctional apparatus for the synthesis and in situ investigations of low-dimensional structures using the spectral magnetic-ellipsometry method in the temperature range of 85–900 K was developed on the basis of the measuring and structure-growing complex (MSGC) for synthesis and in situ investigations of spintronics materials [10]. As a result of its upgrading, the MSGC was additionally equipped with a specially developed and manufactured linear bellows translator, a lock chamber for recharging, a slide gate, and a vacuum bellows three-degree-of-freedom manipulator (*13*), to which a rod with the sample holder is attached (Fig. 1). This figure also shows the system for supplying liquid nitrogen (*18*) that provides the temperature measurement range of 85–500 K. The sample temperature is specified via the liquid-nitrogen supply to a flow-type evaporator–heater (*14*) with the subsequent heating of gaseous nitrogen to a predetermined temperature. The heated gaseous nitrogen is fed to the sample holder through the rod (*2*).

The sample holder (*10*) (Fig. 2a) is a copper base on which a sample (*13*) is mounted through dielectric sleeves (*3*) using screws (*1*) and contacts (*5*). The sample holder (*10*) is connected to the rod of coaxial design using the argon arc welding technique. The upper end of the rod is connected to the vacuum bellows inlet using the argon arc-welding technique (*13* in Fig. 1). A sample is installed on a dielectric sapphire plate, which serves as the electric insulator and heat conductor at low temperatures. The sample holder allows mounting of substrates with a width of 10 mm, a length of 35 mm, and a thickness of at most 3 mm. The sample-holder temperature monitoring at temperatures of 85–500 K is provided by a ChEP-2888 platinum thermistor (*8* in Fig. 2a). Figure 2b shows a schematic diagram of the sample holder combined with a mounted sample.

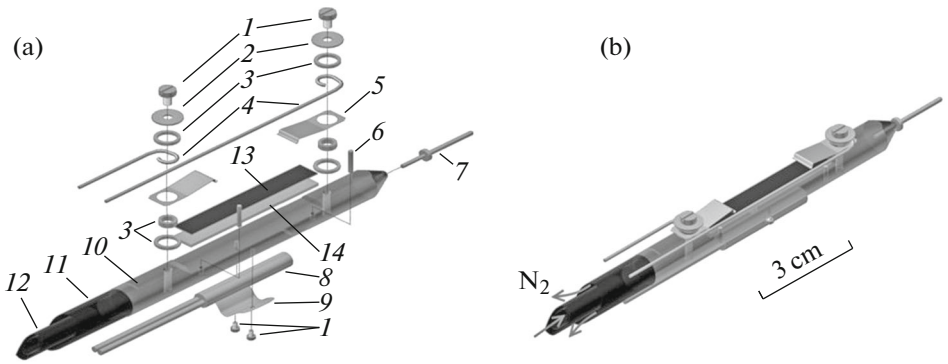

Fig. 2. The sample holder design: (*1*) screw; (*2*) washer; (*3*) dielectric sleeves; (*4*) current lead; (*5*) contacts; (*6*) stoppers; (*7*) stopping element; (*8*) ChEP-2888 platinum thermistor; (*9*) attachment; (*10*) sample holder; (*11*) rod; (*12*) coaxial tube; (*13*) sample; and (*14*) dielectric plate.

Tantalum contacts (*5*) (Fig. 2a) also serve as the electrodes in the sample heating system, which provides a temperature range of 300–900 K. Electric current leads (*4*) are used to pass an electric current and heat the sample, which are attached to the sample holder in its upper and lower parts using screw clamps (*1*). The sample temperature is monitored in the range of 570–900 K with an IMPAC IGA 5 single-beam pyrometer (LumaSense Technologies) with a fixed coefficient of gray of 0.5.

TESTS OF THE ULTRAHIGH-VACUUM MULTIFUNCTIONAL APPARATUS

To test the ultrahigh-vacuum multifunctional apparatus and the created system for setting the sample temperature, experiments on determining the complex permittivity of Si at different temperatures were performed in this study using an Ellipse-1891 fast spectral ellipsometer (produced at the Rzhanov Institute of Semiconductor Physics, Novosibirsk) [11]. Single-crystal silicon was chosen as an experimental sample because there is a large amount of experimental data for it [8, 12–16].

Despite the possibility of direct measurements of the silicon permittivity at various temperatures in an ultrahigh vacuum using the Si surface that was cleared of oxides, most papers are devoted to measurements of the optical properties of the Si surface with an artificially formed thin SiO_2 layer on it. This is caused by a change in the morphology of the surface of pure single-crystal silicon under high-temperature heating of a sample [15]. An artificial layer of silicon dioxide with a thickness of several nanometers reduces the probability of the formation of islands on the SiO_2–Si interface. Therefore, it is unnecessary to create complex optical models of a transition rough layer, which may significantly distort the results of ellipsometric measurements.

To evaluate the reliability of in situ spectral ellipsometric measurements in the temperature range of 85–900 K, an experiment with a single-crystal silicon substrate, which was coated with a SiO_2 buffer layer, was performed on the described apparatus. A KDB 5-15 (100) single-crystal silicon wafer (10 × 30 × 0.3 mm) served as the experimental sample. A SiO_2 layer with a thickness of 26 nm was formed on the sample surface using the method of thermal oxidation in the O_2 + HCl medium at the temperature of 1270 K. To reduce the number of defects in the deposited oxide layer, the substrate was annealed for 15 min in an inert atmosphere of highly pure nitrogen. Contact pads with a thin SiO_2 layer (~1-nm thick) were etched out at the sample edges to provide its heating via a direct current passage. The etching procedure was performed by immersing the sample in a 10% HF solution for a short time at room temperature with the preliminary deposition of a polyvinylchloride mask on the central part of the sample. After being etched, the sample was washed in deionized water and the mask was removed.

Before being charged into the apparatus, the sample was additionally cleaned in a boiling solution of NH_4OH (1 part) : H_2O_2 (1 part) : H_2O (1 part) for 20 s and then finally washed in deionized water and dried in air.

The SiO_2 thickness was measured in air in the central part of the sample immediately before its charging using a fast Ellipse-1891 spectral ellipsometer in the four-zone mode [11, 17]. The optical modeling of the "vacuum–SiO_2 layer–Si substrate" system showed the presence of an oxide layer with a thickness of 25.70 ± 0.06 nm.

After the sample was charged into the ultrahigh-vacuum chamber, its position relative to the optical system of the in situ spectral ellipsometer was adjusted so as to provide the coincidence of the measured SiO_2 thickness on the sample with the previously measured value in air before charging. Thus, the light angle of incidence with the normal to the sample surface was

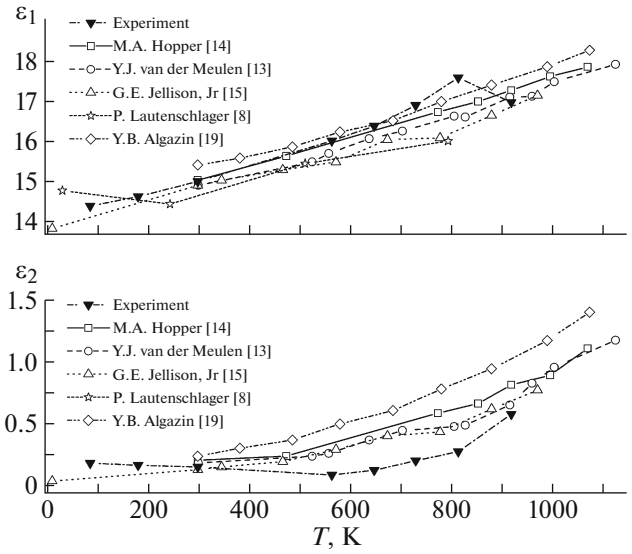

Fig. 3. The temperature dependences of the complex permittivity of Si at a wavelength of 632.8 nm (1.96 eV) that was calculated from the results of ellipsometric measurements.

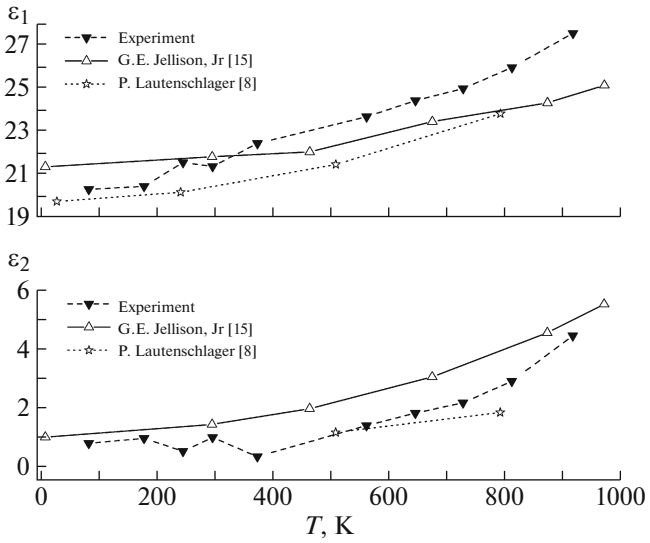

Fig. 4. The temperature dependences of the complex permittivity of Si at a wavelength of 459.3 nm (2.70 eV) that was calculated from the results of ellipsometric measurements.

55.90° [3]. Subsequently, the sample surface was degassed in an ultrahigh vacuum at $T = 600$ K. The sample was annealed via a direct current flow (0.2 A) through the sample under the control of the residual-atmosphere pressure in the technological chamber (no worse than 10^{-6} Pa) for 1 h.

To determine the permittivity of bulk silicon, spectral ellipsometric measurements were performed on the prepared sample at different temperatures between 85 and 900 K. The measured spectral dependences of the ellipsometric parameters were then converted into the spectra of the complex permittivity $\varepsilon = \varepsilon_1 + i\varepsilon_2$ for the substrate that was under the SiO_2 layer with a thickness of 25.7 nm. The permittivity ε was calculated and fitted using the single-layer ellipsometric model [3] and the Nelder–Mead simplex-optimization method [18]. The temperature variations in the SiO_2-layer permittivity were disregarded in the calculation as they were small [13] in comparison with the error of ellipsometric measurements, which is at most 1% of the measured quantity.

To compare the measurement results with data of other researchers, we used the method of selective storing of ellipsometric signals at the following points in the spectrum: 632.8 nm (1.96 eV) and 459.3 nm (2.70 eV). The temperature dependences of the calculated ε_1 and ε_2 for silicon were compared with the measurement results of other authors (Figs. 3 and 4).

As a whole, good agreement of the silicon-permittivity values that were found in this study with the results from [8, 13–15, 19] is observed in the temperature range of 85–900 K at wavelengths of 632.8 and 459.3 nm.

CONCLUSIONS

The design of the ultrahigh-vacuum multifunctional apparatus with a built-in in situ spectral magnetic ellipsometer, a system for setting the sample temperature in a wide range, and an evaporation system on the basis of Knudsen cell makes it possible to perform high-temperature annealing of semiconductor and metal substrates, synthesize thin films on their surfaces, and measure the dependences of the optical and magneto-optical properties of the produced structures at different temperatures in the range of 85–900 K.

In tests of the ultrahigh-vacuum multifunctional apparatus, the temperature dependences of the spectrograms of the bulk-Si complex permittivity were measured and the results were compared with the literature data. Hence, it has been shown that this technological apparatus can be widely used in materials science and investigations of solid-state reactions and various phase transitions.

ACKNOWLEDGMENTS

This study was supported by a Grant of the President of the Russian Federation for Support of Leading Schools (project no. NSh–7559.2016.2), by the Russian Foundation for Basic Research, the Government of Krasnoyarsk krai, and the Krasnoyarsk krai foundation for support of scientific and research and technical activity as a part of the scientific project no. 16-42-243058.

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Translated by A. Seferov