

Measurements of Nonstationary Temperatures by the Spectral Pyrometry Method

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Received June 30, 2011

Abstract—Recording of a sequence of thermal-radiation spectra allows determination of a nonstationary temperature $T(t)$ without using the data on the emissivity of an object. For a КЭФ-4.5 silicon single crystal heated with radiation from a continuous-wave Nd:YAG laser ($\lambda = 1.064 \mu\text{m}$), sequences of hundreds of emission spectra in wavelength ranges of $\lambda = 350\text{--}760 \text{ nm}$ and $\lambda = 650\text{--}1000 \text{ nm}$ were recorded at a signal storage time of a CCD array of $\tau = 15\text{--}35 \text{ ms}$ and a frequency of recording spectra of $f \approx 30\text{--}66 \text{ Hz}$. The spectra were automatically processed, and the dependences of the crystal temperature on the time after the irradiation onset were obtained in the range $T \approx 1100\text{--}1450 \text{ K}$.

DOI: 10.1134/S0020441212010095

INTRODUCTION

Nonisothermal methods are widely used to study the kinetics and mechanisms of physicochemical processes in inorganic [1–4] and organic [5] chemistry, laser thermochemistry [6, 7], and plasmachemistry [8–10]. In a nonisothermal regime, processes occur when the system contains an internal (chemical) or external (laser, plasma, microwave, ion- or electron-beam, current) heat source and heat-removal channels do not provide thermal stabilization. Under such conditions, it is necessary to measure the nonstationary temperature $T(t)$, the differentiation of which allows one to determine the temperature dependence of the power $P \sim dT/dt$ of heat sources that heat and cool an object.

In the region of high temperatures ($T \geq 1000 \text{ K}$), temperature measurements are performed using optical-pyrometry methods (OPMs). In this case, the main problem is that there are no data on the emissivity of many materials.

In recent years, the OPM has been underway [11], in which the absence of data on the emission properties of an object is compensated by extensive spectral information. A CCD spectrometer is used to record a wide radiation spectrum, the verification of its similarity to a Planck spectrum is performed, and then the temperature is calculated using the intervals where the spectra are similar. As a rule, in the studied process, a single spectrum is recorded, from which one temperature value averaged over the signal storage time is calculated. However, this is often insufficient for describing this process.

In this study, we recorded sequences that consisted of 300–1500 spectra at a spectrum-recording frequency of 30–66 Hz and constructed the dependences

of the temperature on the time that passed from the heating onset of a silicon single crystal irradiated by a laser beam. The characteristics of the setup and the possibilities of the automated processing of spectra for temperature calculation are discussed.

EXPERIMENT

The setup includes a continuous-wave Nd:YAG laser ($\lambda = 1.064 \mu\text{m}$) with a power of up to 30 W, a power meter, a sample holder, and HR 2000+ fiber-optic CCD spectrometers (Ocean Optics) for recording spectra. Spectra are displayed on a computer monitor, to which one or several spectrometers can be connected, and all spectra are observed simultaneously. The use of several spectrometers makes it possible to extend the wavelength range for recording spectra and compare the results obtained in different spectrum regions. The silicon array of each spectrometer consists of 2048 pixels.

The relative spectral sensitivities of the spectrometers are calibrated using an LS-1-CAL standard tungsten lamp whose spectrum is known from the results of measurements that were performed at a metrological laboratory. The primary radiation spectrum of any object measured by a spectrometer contains the dependence of the number of photoelectrons in a pixel on the wavelength that corresponds to a given pixel. Then, the primary spectrum is automatically corrected by multiplying the signal of each pixel by the calibration coefficients $k(\lambda)$, which are defined as the ratio of the intensities in the actual spectrum of the standard lamp to the intensities in the spectrometer-recorded spectrum of the same lamp. The thus corrected spectra are expressed in the units of the intensity ($I, \mu\text{W}/(\text{cm}^2 \text{ nm})$) that is incident on the input of

the optical fiber. The two spectrometers used in our measurements are calibrated in the wavelength ranges $\lambda = 350\text{--}760\text{ nm}$ and $\lambda = 650\text{--}1000\text{ nm}$, although the spectrometers allow the recording of wider wavelength ranges (the operating wavelength ranges for these spectrometers are $350\text{--}800$ and $650\text{--}1080\text{ nm}$). The spectral width corresponding to each pixel in the CCD array is 0.2 nm .

The spectrometer program allows one to specify the storage time of an individual spectrum, the time interval between neighboring spectra, and the total recording time of the spectra (or their number). The spectrum recording time of the CCD spectrometer is determined by the storage time, which depends on the brightness of the object and the distance to it. The storage time of individual spectra of the CCD array is chosen in an interval of $1\text{--}20\text{ s}$ and is determined by the trial and error method for each object and measurement conditions so as to attain an intensity that suffices for further processing but does not reach the signal saturation level or its cutting-off during digitization.

A program that allows representation of each spectrum on the coordinate plane (x, y) , where $x = \ln(\lambda^5 I)$ and $y = C_2/\lambda$, was developed for determining the temperature from radiation spectra. In these coordinates, a spectrum can be approximated by the Wien function (the short-wavelength of the Planck function). The equation of the straight line that approximates an experimental spectrum (or its segment) in this coordinate system has the form

$$y = A - Tx,$$

where A is a constant and T is the sought temperature of the object.

The spectral region over which the temperature must be calculated is selected visually. In this case, one has to exclude the short-wavelength edge of the experimental spectrum where the signal is small and very noisy. In addition, the wing of a scattered laser line is observed at the long-wavelength edge of the spectrum in the spectrometer operating in the wavelength range of $650\text{--}1000\text{ nm}$. Therefore, the wavelength range with a width of $10\text{--}20\text{ nm}$ near $\lambda = 1000\text{ nm}$ is not used in calculations.

In many studies, spectrometers are calibrated not in power units but in units that are proportional to the number N of photoelectrons in pixels [11]. In this case, the values $x = \ln(\lambda^4 N)$ are plotted along the abscissa axis.

Figure 1 shows the window of a program that allows calculation of the temperature of a specimen from its emission spectrum. Files that contain recorded digitized spectra are written in the program memory. Files are sequentially called by the program. The number of spectra may range from several units to several tens of thousands. The wavelength range for calculating the temperature is set by two markers. After the interval within which the temperature must be calculated is

chosen, the unified calculation is performed for all spectra from the series stored in the program memory.

The time during which a sequence of spectra can be recorded is limited only by the computer memory capacity (e.g., a sequence consisting of 1300 spectra within a wavelength range $\lambda = 650\text{--}1050\text{ nm}$ has a size of 50 MB).

RESULTS AND DISCUSSION

Figure 2 shows the spectra of thermal radiation from a flat specimen of a $K\Theta\Phi\text{-}4.5$ silicon single crystal (electron conduction, the doping impurity is phosphorous, the electric resistivity is $4.5\ \Omega\text{ cm}$). The 0.4-mm -thick specimen is irradiated from above perpendicularly to the polished surface with a laser beam with a power density of 100 W/cm^2 in a multimode regime. The spectra were recorded with intervals of 1 s in two different experiments. The size of the heated specimen ($2 \times 2\text{ mm}^2$) is smaller than the beam diameter ($\approx 3\text{ mm}$). The crystal lies on a 0.3-mm -thick polycor substrate (corundum-based ceramics), which is transparent for laser radiation. The polycor melting temperature is higher than 2000 K . The receiving ends of 0.6-mm -diameter optical fibers are positioned at a distance of 4 cm from the specimen.

Figure 3 shows the same spectra in the Wien coordinates. The temperature is determined from the slope of the curves that approximate the chosen segment of the experimental spectrum. The equation of each curve and the desired temperature is determined by the least-squares method. The correlation coefficient between the experimental points and the straight line is usually no less than 0.995 . The spectra with substantial deviations from a rectilinear shape are rejected.

Figure 4 shows the time dependences of the crystal temperature after switching the laser on and off for two pumping-power levels ($P = 12$ and 16 W , the power density on the specimen $D \approx 130$ and 180 W/cm^2 , respectively). The large number of points on each curve allows numerical differentiation and evaluation of the heating and cooling rates dT/dt . The observed crystal heating rate at $T \geq 1100\text{ K}$ reaches $1000\text{--}1500\text{ K/s}$, and the cooling rate after switching the laser off reaches $3000\text{--}5000\text{ K/s}$ (estimated values).

As is seen, at a higher pumping level, a steady-state temperature is reached as a result of an abrupt decrease in the heating rate. This may be associated with a nonlinear decrease in the absorption coefficient of the incident power (e.g., owing to an exponential increase in the concentration of free carriers in a crystal) or a rapid increase in the heat loss (the power of heat removal by radiation is proportional to $\sim T^4$). The integral heat losses at any temperature that include, apart from the heat removal by radiation, the heat removal to the substrate are determined on the basis of the crystal cooling rate after switching the laser off.

For radiation of the first harmonic of the neodymium laser, the silicon single crystal at $T = 300\text{ K}$ is

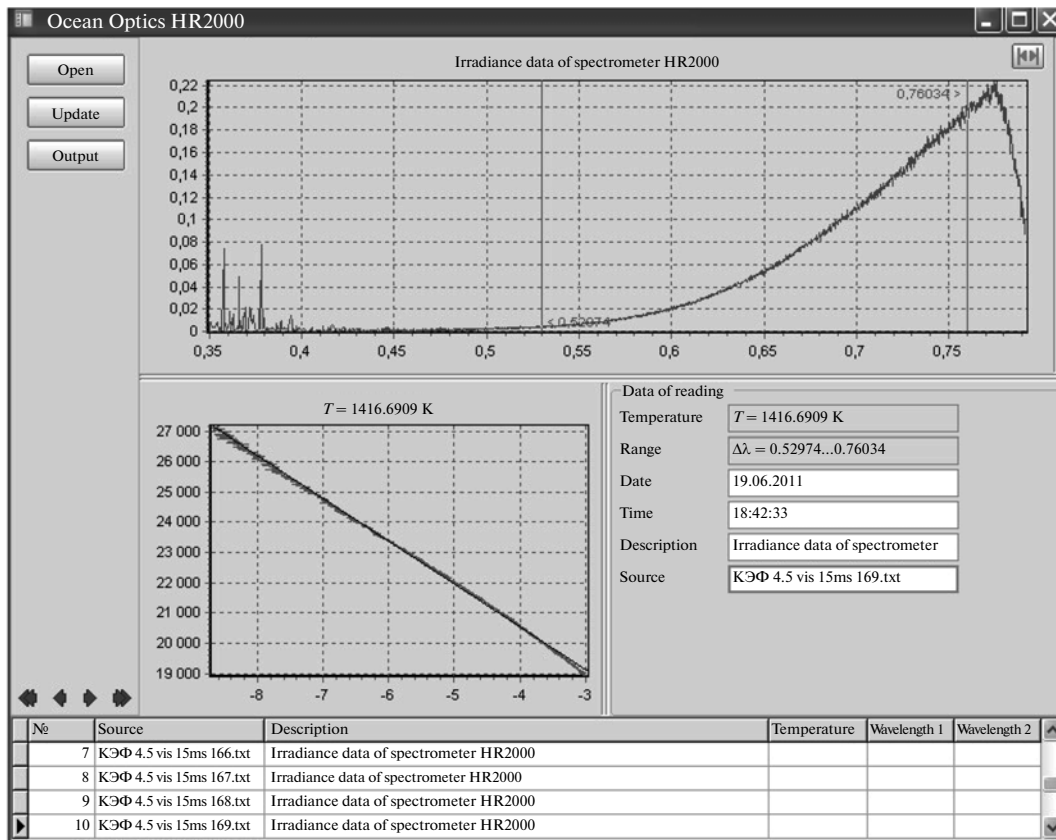


Fig. 1. Window of the “Spectral Pyrometry” program. The temperature of the silicon single crystal is calculated from the thermal spectrum within a wavelength range of $\lambda = 530\text{--}760$ nm (upper window). The selected spectral fragment (lower window) is constructed in the Wien coordinates, the slope of the straight line is governed by the temperature ($T \approx 1417$ K). The spectrum storage time is 15 ms.

semitransparent (the linear absorption coefficient $\alpha \approx 10 \text{ cm}^{-1}$); at a thickness of 0.4 mm, the light transmission coefficient for the crystal is approximately 0.3. Therefore, when the specimen is heated, the laser light is absorbed in the bulk almost uniformly. As the temperature rises, the edge of the band-to-band light absorption shifts toward longer wavelengths; thus, the absorption coefficient increases, and at $T > 570$ K, the crystal becomes nontransparent for the first harmonic of laser radiation. Therefore, at higher temperatures, heating loses the bulk character and acquires a surface character (at $T = 1000$ K, $\alpha \approx 2500 \text{ cm}^{-1}$); thus, radiation is mainly absorbed in a surface layer with a depth smaller than $h \approx 1/\alpha \approx 4 \text{ }\mu\text{m}$.

Temperatures below 1100 K cannot be determined using silicon CCD arrays at the chosen storage time (15 ms) of the visible spectrum because of a low signal level. Temperatures exceeding 1000 K can be determined at the same storage time using a spectrometer of near-IR spectra. If the spectrum storage time in a range of 760–990 nm increases to 35 ms, emission spectra for silicon can be recorded at $T \geq 950$ K. However, this approach does not allow one to advance toward lower temperatures because the radiation

intensity at any wavelength in the region where the Wien law is efficient exponentially drops with a decrease in the object temperature. The ratio of the times that are necessary for storing a signal in pixels and correspond to the same wavelength but different temperatures of the object is defined by the formula

$$t_1/t_2 = [\exp(C_2/\lambda T_1) - 1] / [\exp(C_2/\lambda T_2) - 1].$$

For storing an identical signal at temperatures of 1000 and 900 K, the time ratio is $t_1/t_2 \approx 25$ at $\lambda = 500$ nm and $t_1/t_2 \approx 6$ at $\lambda = 900$ nm.

When the laser power increases to 25 W (at a pumping power density on the specimen of 280 W/cm^2), the melting temperature can be reached, and in this case, the thermal radiation intensity first drops by a factor of almost 2 in the entire spectrum, because the emissivity of liquid silicon at $T \approx 1690$ K is approximately twice as small as that for a solid single crystal at $T \approx 1680$ K [12, 13]. The illumination of the specimen with such a power for several seconds leads to an increase in the temperature to a value of >2500 K, which is accompanied by boiling and spraying of silicon. After the silicon surface is cooled, it stops being flat and becomes irregular. During the next heating of the same specimen, the recorded emission spectrum can be rectified

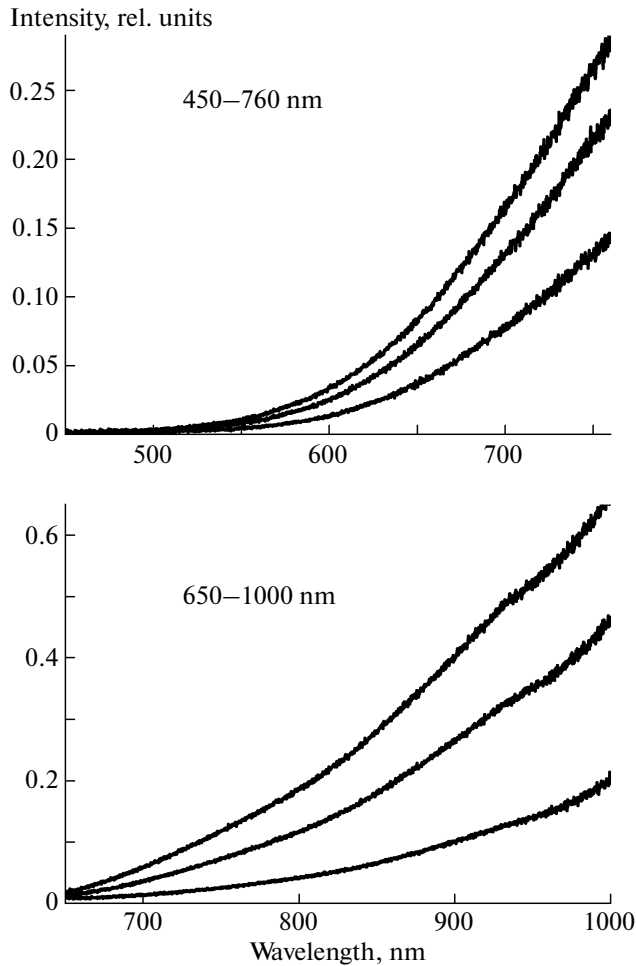


Fig. 2. Spectra of thermal radiation of the silicon single crystal recorded in two spectral ranges with an interval of 1 s under laser heating (the lower and upper spectra correspond to time moments of $t = 1$ and 3 s after the start of heating, respectively). The spectrum storage time is 15 ms, the laser-radiation power density is 100 W/cm^2 .

in the Wien coordinates, thus allowing the determination of the temperature of the rough surface.

However, we failed to select the irradiation conditions under which the temperature would have reached the melting point at a low heating rate ($dT/dt \approx 0$) and would have remained constant for the time required for melting the entire crystal. Estimates show that the time required for melting the crystalline specimen is almost twice as long as the heating time from $T = 300 \text{ K}$ to the melting point (the supplied power being the same). As the laser power gradually increased, a significant temperature increase was observed (from $T \approx 1550 \text{ K}$ to values that are 100–200 K higher than the melting point $T_m = 1685 \text{ K}$). In this case, large chaotic temperature oscillations were observed. The main channel of heat losses is the heat removal to the substrate, which is 2–4 times higher

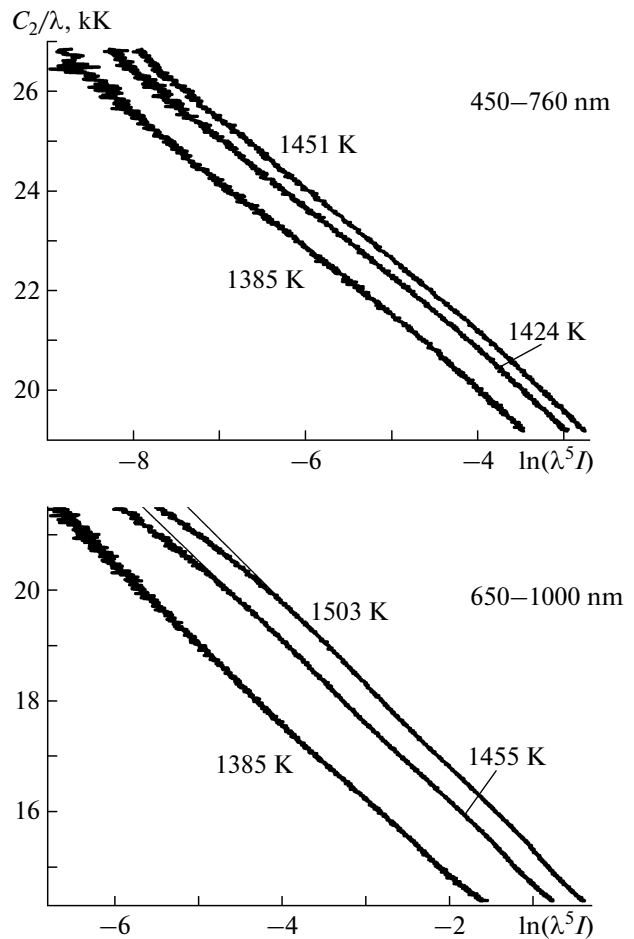


Fig. 3. Spectra of Fig. 2 in the Wien coordinates (the lower and upper spectra at $t = 1$ and 3 s, respectively). The slopes of straight lines are determined by the temperature indicated near the curves.

than the radiation loss, and the thermal resistance of the specimen–substrate contact is reproduced poorly.

The search of a regime with temperature stabilization under continuous irradiation of the crystal will be continued because the achievement of the melting point and recording of the segment $T(t) = \text{const}$ makes it possible to determine the systematic error of measuring the temperature from the emission spectrum.

Thus, the thermal spectra of a Si single crystal at temperatures of >1000 – 1100 K and the spectra of a Si melt at $T \geq 1685 \text{ K}$ are reliably recorded by silicon CCD arrays. The possibility of measuring temperatures within a range of 2500 – 8000 K from the emission spectra of various objects (condensed media, small particles) was demonstrated in several papers [14–16], but the problem of recording a large number of spectra and determining a nonstationary temperature was not posed previously.

In this study, we did not reach the maximum frequency of recording spectra, whose value for the chosen type of CCD arrays is 1000 Hz at a minimum spec-

trum storage time of 1 ms, because the brightness of the studied single-crystal silicon specimen is insufficiently high. Near the crystal melting point, the radiation intensity is high and close to the saturation of the CCD spectrometer at the chosen storage time and the geometry of the experiment. At this temperature, a considerable increase in the spectrum-recording frequency can be observed with a decrease in the storage time. For example, during laser heating and burning of Ti and Fe microparticles in air, spectra can be stored within 3–5 ms and frequencies of recording of 200–330 Hz can be reached. An experimental problem is that it is desirable to attain the highest resolution at comparatively low temperatures and a low intensity of thermal radiation, i.e., at the heating stage where the rate dT/dt is high. It is obvious that, when spectra are stored, the temperature must not change to a significant degree; otherwise, only the most intense radiation will be detected within a short period when the temperature is especially high.

CONCLUSIONS

Spectral pyrometry allows measurements of the nonstationary temperature of a single crystal heated with laser radiation regardless of the phase state of its surface (below or above the melting temperature) and the surface state (polished or rough). Recording of a sufficiently wide spectrum of thermal radiation and its similarity to the blackbody spectrum is a sufficient condition for determining the crystal temperature. The similarity of these spectra is not postulated beforehand but is checked in each experiment: to what degree the recorded spectrum is rectified in the coordinates of the Wien law. In this study, we attained a time resolution of 15 ms, which is determined by the storage time of spectra in the CCD array. In this case, the crystal temperatures were determined in the range of ≈ 1100 – ≥ 2500 K. The obtained dependences $T(t)$ include 200–1300 points, each of which was obtained via processing of a single emission spectrum. At comparatively low laser powers when the silicon melting point is not reached, the dependences $T(t)$ are monotonic. These results create a methodological basis for studying nonisothermal high-temperature regimes and features of the heat exchange under optical heating of single crystals in fast thermal processes in microtechnologies.

The performed investigation demonstrates both the prospects of the use of spectral pyrometry for recording nonstationary temperatures and the limited capabilities of universal spectrometers for solving this problem. The use of thermal optical spectra, in which the radiation intensity increases exponentially with both the wavelength and the temperature, in pyrometry requires specially arranged optical and electronic schemes of pyrometers. One spectrometer with a fixed wavelength range does not allow recording of the entire heating kinetics (an increase in the intensity at a

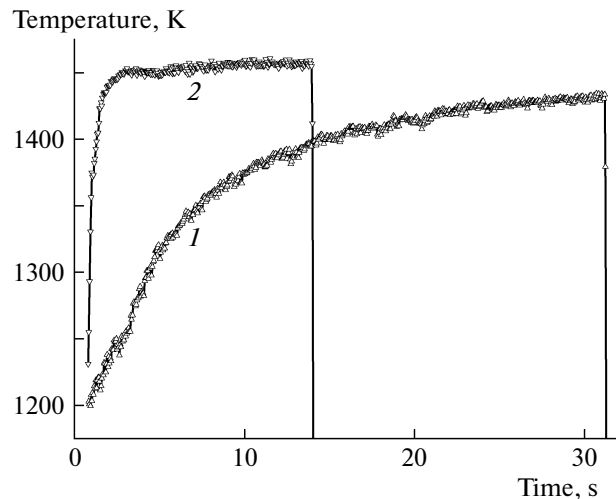


Fig. 4. The dependences of the temperature of the 0.4-mm-thick silicon single crystal on the time after switching the laser on and off for two pumping-power levels P at power densities D on the specimen: (1) $P = 12$ W, $D = 130$ W/cm²; (2) $P = 16$ W, $D = 180$ W/cm².

wavelength of 760 nm within the entire dynamic range of a photodetector corresponds to a temperature interval of 1000–1600 K). In order to record spectra in a wide temperature range, radiation should be detected using several spectral pyrometers, which are connected in parallel and encompass a wide wavelength range. As the temperature increases, the spectral segments used for its calculation should be shifted toward shorter wavelengths. Silicon CCD arrays whose sensitivity is restricted to a wavelength range of $\lambda = 200$ –1080 nm are unsuitable for measuring temperatures below 1000 K.

In order to record the dependence $T(t)$ beginning with room temperature, an additional spectrometer with an array of photodetectors, which are sensitive to radiation with wavelengths of 1–3 μm , is necessary. By now, spectral pyrometers with high sensitivities in wavelength ranges of $\lambda = 1.3$ –1.6 μm for measuring temperatures $T \geq 500$ K [17] and $\lambda = 1.2$ –3.4 μm for measuring temperatures $T \geq 400$ K [18] have been developed.

Studying nonstationary processes during laser heating of materials requires recording of thermal-radiation spectra within a time of several microseconds. To solve this problem, it is necessary not only to increase the response speed of CCD arrays but also to considerably enhance the spectrometer luminosity, which is insufficiently high when using a fiber-optic input in the absence of gathering optics.

ACKNOWLEDGMENTS

We are grateful to N.I. Timchenko for the development of the program for processing spectra and calculating the temperature.

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