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SPECTRORADIOMETRY OF ULTRAVIOLET RADIATION

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The principles underlying the construction of primary standard detectors and sources of ultraviolet radiation are considered. A double ionization chamber, proportional counter, and cryogenic radiometer with electrical displacement are used as the standard radiation detectors while synchrotron radiation the spectral characteristics of which are described on the basis of Schwinger's theory serves as the standard source. Results of international key comparisons that confirm the high accuracy of the methods and instruments that have been developed at the All-Russia Research Institute of Optophysical Measurements (VNIIOFI) are presented.
Keywords: *synchrotron radiation, ionization chamber, electron storage ring, proportional counter, cryogenic radiometer, primary standard detector.*

Measurements of the spectroradiometric characteristics of radiation sources and detectors serve as a basis for the solution of problems in the radiometry of extremal vacuum and near-ultraviolet radiation in ecology and medicine, space exploration, and the photochemistry and diagnostics of plasma. The development of the spectroradiometry of ultraviolet radiation has been associated with the monitoring of solar activity, the study of processes of photoionization in the upper layers of the atmosphere, the creation of systems for the detection and ranging of plasma objects, and studies in the area of nanophotolithography, which has become one of the most promising trends in the field of nanoelectronics.

The spectroradiometry of near-ultraviolet radiation in the range of wavelengths 200–400 nm is used particularly often for fundamental medical and biological studies in cytology, immunology, and oncology; for phototherapy, monitoring the effectiveness of bactericidal irradiators and solariums, the creation of solar simulators, and monitoring the safety of working conditions. The range of extremal vacuum ultraviolet radiation in the range 1–40 nm became accessible for extensive studies in recent decades once the difficulties characteristic of this range and associated with the low efficiency of the reflecting coatings of mirrors and diffraction gratings, the need to perform adjustments of optical systems under the conditions of a vacuum, the absence of transparent materials for the fabrication of mirrors of sources and detectors of radiation, and the degradation of the elements of spectral and polarization devices under the action of jets of erosive plasma that accompany the radiation of open sources had been overcome.

A new approach based on the use of a single-electron regime in modern circular-orbit accelerators and the application of highly sensitive cooled charge-coupled arrays with surface nanostructures has been implemented in recent years at national metrological institutes for use in the spectroradiometry of ultraviolet radiation. The approach yields an increase in the accuracy of reproduction and dissemination of units of the corresponding quantities, inasmuch as it makes it possible to relate the characteristics of the synchrotron radiation of circular-orbit accelerators to the fundamental physical constants. Studies on metrological assurance in the field of extremal vacuum ultraviolet radiation for nanoelectronics, diagnostics of thermonuclear plasma, and space-based monitoring of solar activity are among the principal trends of optical radiometry and are carried out with the use of the MLS, BESSY-II and SURF-III, and NSRS modern electronic storage rings. The basic problems in the

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spectroradiometry of ultraviolet radiation are related to the need to create methods and instruments for the dissemination of the unit of irradiance of radiation with high spatial resolution, and the creation of methods of fabricating high-efficiency reflecting coatings and mirrors for use in segregating spectral ranges. Such studies have also been carried out at the national metrological institutes of Japan, France, and China.

Methods of plasma diagnostics based on an analysis of the spectro-zonal images of plasma objects require absolute calibration of the comparator, a device which comprises an image generation system and a positionally sensitive charge-coupled detector. In the ultraviolet range, the use of charge-coupled arrays is limited by the nontransparency of the conducting polycrystalline radiation shutters with wavelength shorter than 400 nm. Charge-coupled arrays with reverse exposure are used in the region of near and vacuum ultraviolet radiation. The image of the radiating region is produced on the reverse side of a parallel register lacking any shutters.

The development of microelectronics is characterized by a transition to nanoelectronics with the use of vacuum ultraviolet radiation. Recent advances in photolithography have made possible a dimension of the elements less than 30 nm due to the use of extremal vacuum ultraviolet radiation with wavelength close to 13.5 nm for the exposure of photoresists. This requires the creation of a system of real-time control of the absolute spectral sensitivity of the photoresists with the use of standard detectors in a given region. Another important field, which is responsible for the development of the spectroradiometry of extremal ultraviolet radiation, is the diagnostics of the temperature and concentration of high-temperature plasma.

The most common primary standard detectors in this area are ionization chambers and proportional counters. A double ionization chamber is a primary standard detector in the area of vacuum infrared radiation, where only first-order ionization of the atoms of an inert gas is possible. A proportional counter is used at high photon energies in the area of extremal ultraviolet radiation, when there is multiple ionization, the quantum efficiency or absolute spectral sensitivity of which is calculated based on the energy of first-order ionization, the electron charge, and the gas-amplification factor. Ionization chambers and proportional counters function in a permanent regime that is most successfully used in combination with monochromatic synchrotron radiation. In this case, the spectral flux of photons is recorded in a dynamic range of several dozen orders of magnitude as a consequence of the variation of the current of the electron storage ring. Ionization chambers and proportional counters with known quantum efficiency are used to determine the absolute spectral sensitivity of semiconductor photodiodes. The main advantages of a proportional counter are the possibility of using it to measure fluxes of photons of high energies, high spectral sensitivity, and high time resolution.

An ionization chamber is filled with an inert gas the quantum photoionization yield of which is equal to 1. Absorption of photons in the ionization chamber is determined by the Bouguer–Lambert–Beer law:

$$N = 1 - N_0 \exp(-\sigma n_0 l); \quad N_0 = N[1 - \exp(-\sigma n_0 l)]^{-1},$$

where N and N_0 are the number of photons absorbed in the ionization chamber and entering the chamber in unit time, respectively; σ, absorption cross-section; n_0, number of molecules in a unit of volume; and l, length of the ionization chamber.

The double ionization chamber method, by means of which the gas pressure may be eliminated from the analysis, is used to determine the number of photons. In such a chamber, the number of absorbed photons in the first section N' is determined by the expressions

$$N' = N_0 \exp(-\mu d)[1 - \exp(-\mu L_1)]; \tag{1}$$

$$\mu = \sigma n_0 = \ln(i_1/i_2)/d, \tag{2}$$

where L_1 and d are, correspondingly, the dimensions of the ion collectors of the first section and of the guard ring at the entrance to the chamber, and i_1 and i_2 the ion saturation currents of the collectors of the first and second sections of the chamber.

The number of absorbed photons in the second section N'' is calculated thus:

$$N'' = N_0 \exp(-\mu L_1 - \mu d)[1 - \exp(-\mu L_2)].$$

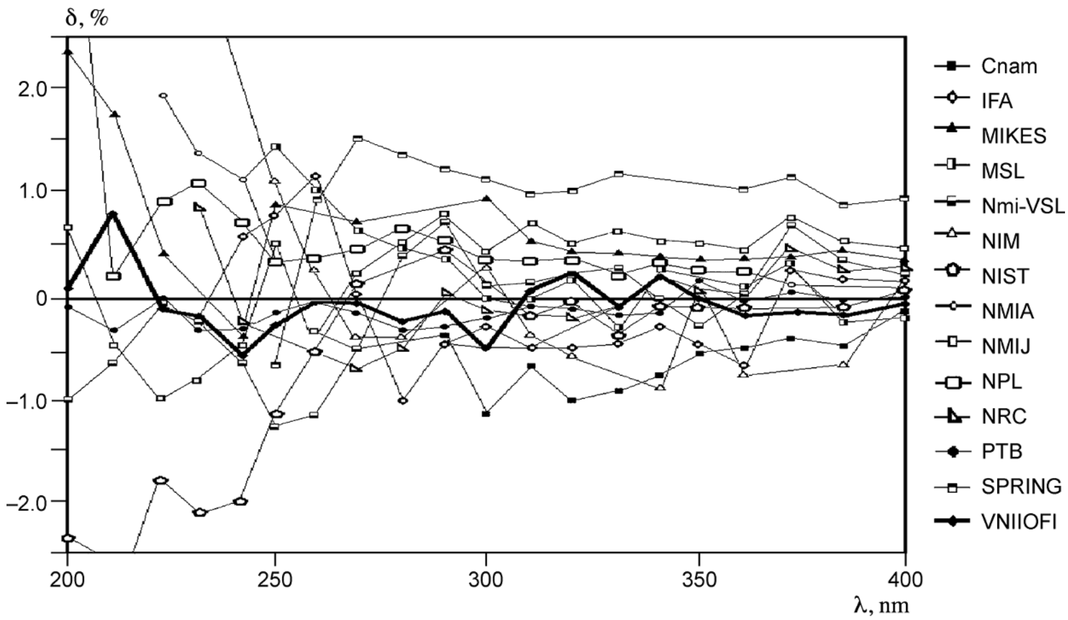

Fig. 1. Deflections from reference values of results of measurements of the absolute spectral sensitivity of photodiodes.

From (1) and (2) with $N' = i_1/e$, $N'' = i_2/e$, where e is electron charge, we obtain a formula for the reproduction of a radiation flux with the use of a double ionization chamber as the standard detector:

$$\Phi_{\Delta\lambda} = \frac{hc}{e\beta} \int_{\Delta\lambda} \frac{(i_1 + i_2 + i')}{\lambda[1-(i_1/i_2)^{-l/d}]} d\lambda,$$

where h is Planck's constant; c, speed of light; λ, wavelength; β, quantum photoionization yield; and i', ion saturation current in the collector in front of the input diaphragm.

When recording monochromatic radiation with wavelength λ, the ratio between the photocurrent of the secondary standard (SS) i_λ^{SS} and that of the ionization chamber (IC) i_λ^{IC} is equal to the ratio between the values of the quantum yield:

$$i_\lambda^{\mathrm{SS}} / i_\lambda^{\mathrm{IC}} = \gamma_\lambda^{\mathrm{SS}} / \gamma^{\mathrm{IC}},$$

where i_λ^{SS} is the quantum yield of the standard.

The absolute spectral sensitivity S_λ of the secondary standard

$$S_\lambda = e\lambda\gamma_\lambda^{\mathrm{SS}} / (hc\rho_\lambda) = e\lambda i^{\mathrm{SS}} / (hc\rho_\lambda i^{\mathrm{IC}}),$$

where ρ_λ is the spectral reflection factor of the comparator mirror, and

$$i^{\mathrm{IC}} = (i_1 + i_2 + i') / [1-(i_1/i_2)^{-l/d}].$$

A cryogenic radiometer with electrical displacement is used as the standard detector in the spectral ranges of vacuum and near-ultraviolet radiation. Through the use of such a radiometer and radiation source with wavelength tuning, it becomes possible to perform calibration of detectors at an arbitrary wavelength over a broad spectral range. The operating principle of such a radiometer is based on transformation of an incident flux of radiation Φ into heat by means of a cavity with absorption coefficient close to 1. The radiation absorber is equipped with a thermometer for measurement of the temperature T_{a} and a heater, which is connected to a power source. The absorber is linked to a cooler found at a constant temperature T: $T_{\mathrm{a}} - T = R_T\Phi$, where R_T is the thermal resistance.

The electrons and ions formed in the space of a proportional counter as a result of photodissociation experience collisions with the gas molecules that fill the counter as they travel towards the electrodes. The quantum energy of the radiation exceeds the threshold of first-order ionization and an incident photon is led to form an electron – ion pair with charge $2e$, multiplied by the quantum efficiency of the gas and the gas-amplification factor. The voltage across the electrodes is ten times greater than the voltage in the ionization chamber, amounting to more than 1 kV. As the electric field strength E increases, the electrons acquire energy sufficient for ionization of the gas: $W_{ion} = eE\lambda_{ion}$, where λ_{ion} is the mean free path of an electron corresponding to the ionization energy.

The gas-amplification factor

$$m = N/N_0 = \exp(\alpha x_0),$$

where x_0 is the coordinate of the electron, $\alpha \sim p\exp(-bp/E)$ is the coefficient of collision ionization, which is proportional to the gas pressure p, and b is a constant.

The coefficient of collision ionization depends on the field strength at a point with coordinate r. In a cylindrical counter, a thread serves as an anode of radius r_a while the external surface functions as a cathode of radius r_c. The field strength depends on the distance to the axis. The region where the field strength is sufficient for realization of collision ionization occupies a small volume and is concentrated near the anode. We may therefore write

$$E(r) = V_0\ln(r_c/r_a)/r,$$

where V_0 is the difference between the potentials of the anode and the cathode.

A primary standard ultraviolet radiation detector, which comprises a double ionization chamber, a cryogenic radiometer with electrical displacement, and a proportional counter (in the range of wavelengths 0.0004–0.4 μm) supports reproduction of the units of radiation flux in the range 10^{-11}–10^{2} W; irradiance in the range 10^{-7}–10^{3} W/m^2; spectral density of irradiance in the range 10^{3}–10^{11} W/ m^3; and radiant exposure in the range 10^{-8}–10^{-5} J/ m^2 with standard type A uncertainty $(0.2–0.8)\cdot 10^{-2}$ and standard type B uncertainty $(0.3–1.0)\cdot 10^{-2}$.

The following 14 national metrological institutes participated in the K2c international key comparisons related to measurements of the absolute spectral sensitivity of ultraviolet radiation detectors performed by the International Committee of Weights and Measures in Paris: SNAM (France), National Measurement Institute of Australia (NMIA), Centre for Metrology and Accreditation (MIKES, Mittatekniikan keskus, Finland), IFA (Spain), Measurement Standards Laboratory (MSL, New Zealand), National Institute of Metrology (NMI, China), National Institute of Standards and Technology (NIST, United States), National Metrology Institute of Japan (NMIJ), VSL (National Metrology Institute of the Netherlands), National Physics Laboratory (NPL, Great Britain), National Research Council (NRC, Canada), Physikalisch-Technische Bundesanstalt (PTB, Germany), SPRING (Singapore), and VNIIOFI (Russia).

The deflections of the results of measurements of the absolute spectral sensitivity of PtSi-n-Si Schottky photodiodes in the range of wavelengths 200–400 nm from the reference values are presented in Fig. 1. The results confirm the reliability of the methods and instruments that have been developed by the VNIIOFI and that serve as the basis of the primary standards and have enabled the institute the expand the rows of calibration capabilities.

The principle underlying the design of a primary standard source of ultraviolet radiation is based on the use of the classical theory of synchrotron radiation by means of which the characteristics of the synchrotron radiation of circular-orbit accelerators may be related to the fundamental physical constants of charge, electron mass, and speed of light. The spectral density of a flux of synchrotron radiation $P(\lambda, \Psi)$ found within a unit angle of deflection from the orbit plane is described by Schwinger's theory thus:

$$P(\lambda,\Psi) = [27X/(32\pi^3)][e^2c/R^3](\lambda_c/\lambda)^4\gamma^8[1+(\gamma\Psi)^2]^2 \times$$
$$\times\{K_{2/3}^2(\xi) + K_{1/3}^2(\xi)(\gamma\Psi)^2/[1+(\gamma\Psi)^2]\}, \qquad (3)$$

where Ψ is the angle of deflection from the orbit plane; X, number of accelerated particles; R, radius of orbit; γ, relativistic factor; $\lambda_c = 4\pi R\gamma^{-3}/3$, critical wavelength; $K_{1/3}$ and $K_{2/3}$, special Macdonald functions; and $\xi = [\lambda_c/(2\lambda)]\cdot 1 + [(\gamma\Psi)^2]^{3/2}$, independent variable.

Through integration of (3) with respect to the angles of exceedance over the orbit plane with the use of the relationship $E = 300BR$ between the energy of the particles W and the induction of the magnetic field B at a radiating point of the orbit, a relationship may be obtained that connects the basic parameters which determine the realization of a source of synchrotron radiation:

$$P(\lambda) = 0.22B^7R^4Xf(\lambda/\lambda_c), \qquad (4)$$

where $f(\lambda/\lambda_c)$ is a universal function that describes the spectral distribution of the synchrotron radiation of a relativistic electron.

Strong magnetic fields must be used to increase the density P. An analysis of (4) points to the existence of optimal values of the orbital radius and the energy of the particles corresponding to the maximum density at a given wavelength. In strong magnetic fields, the optimal radius of an orbit decreases rapidly with increasing induction, which simplifies the engineering problem, since strong magnetic fields are realized in small spaces. From (4) there follows an estimate of the maximum density, which may be obtained for a value of the induction B in accordance with the expression

$$P(\lambda) = 5.9\cdot 10^{-17}NB\lambda^{-2}. \qquad (5)$$

The efficiency of different sources may be compared by choosing optimal parameters of a specialized source of synchrotron radiation on the basis of (5). Values of the density P that are ten times as great may be obtained by means of strong magnetic fields. Optimization of the parameters of a specialized source relative to the maximum power of synchrotron radiation in a given spectral region leads to a choice of radius R to within several centimeters. The advantages gained with the use of electron storage rings for radiometry are related to the continuous nature of synchrotron radiation as well as the fact that an individual electron on a relativistic orbit may be segregated.

Axial betatron oscillations of the electrons in a cluster exert the greatest influence on the spectral, angular, and polarization characteristics of synchrotron radiation. The axial oscillations of electrons lead to deflections in the direction of the velocity of individual electrons from the plane of an equilibrium orbit by an angle

$$\Psi' = \dot{z}/c = [A_z(n)^{1/2}/R]\mathrm{Sin}[\omega_0(n)^{1/2} + \varphi],$$

where $\dot{z}$ is the velocity of the electrons in the axial direction; A_z, amplitude of axial betatron oscillations; n, rate of decay of magnetic field in the radial direction; ω_0, frequency of accelerating field; and φ, arbitrary phase.

The density P of radiation segregated by the aperture diaphragm of an optical system is determined by a convolution of the angular distribution function of the synchrotron radiation of an electron $P(\Psi)$ and the distribution function of the electrons with respect to the angles of deflection from the orbit plane $f(\Psi')$:

$$P(\lambda) = \int_{-\Psi_0}^{\Psi_0} d\Psi \int_{-\Psi_{max}}^{\Psi_{max}} P(\Psi - \Psi')f(\Psi')d\Psi',$$

where Ψ_0 is the aperture angle of the optical system, and Ψ'_{max} is the maximal angle of deflection of the velocity of an electron from the equilibrium orbit plane corresponding to the amplitude of axial oscillations.

For a source of synchrotron radiation with optimal radius of the orbit, the angular distribution of the electrons proves to be wider than the angular distribution of the intensity of the synchrotron radiation of an individual electron. The ratio of the spectral fluxes of synchrotron radiation at two wavelengths for a cluster of electrons with high axial dimensions is independent of the dimensions of the cluster and may be precisely calculated. An analysis of the variation in the density P of radiation as a function of the variation in the axial dimension of a cluster shows that the influence of an error in the measurement of the function $f(\Psi')$ decreases with increasing axial dimension of the cluster the most rapidly in the shortwave region, where the condition $\Psi' \gg \Psi_0$ is satisfied the most completely, i.e., the axial dimensions of a cluster of electrons must be increased in order to increase the precision of reproduction of the units. A decrease in the rate of decay, i.e., the gradient of the fundamental magnetic field of the synchrotron in the axial direction in accordance with the expression

$$A_z = c / (\omega_0 (\chi\gamma)^{1/2})[3eE_0 / (m_0 c\omega_0)]^{1/3},$$

where E_0 is the amplitude of the strength of the accelerating UHF field; m_0, rest mass of electron; and χ, rate of decay of magnetic field in the axial direction, proves to be the most efficient method of increasing the amplitude of axial betatron oscillations.

Increasing the axial dimensions of a cluster enables us to overcome the principal drawback of synchrotron radiation, i.e., the angular inhomogeneity of the irradiance. A specialized source of synchrotron radiation may also function in a point cluster regime. It is especially important to achieve angular homogeneity of the ratio of the intensity of the polarization components of synchrotron radiation in precision dissemination of the units of the spectroradiometric quantities of the secondary standard with the sources of unpolarized radiation. The regime of a large cluster supports a constant ratio of the polarized components of synchrotron radiation within a wide aperture angle. The number of particles is determined by a method of comparison of the intensity of synchrotron radiation in a single-electron regime and in the regime of a cluster of electrons with the use of charge-coupled devices that perform generation, accumulation, transmission, and recording of the charge. Through the use of a shutter, it is possible to record a dark frame and to regulate the exposure time. Formers of clock pulses and an analog-to-digital transducer are connected to a charge-coupled array. Random reading noise and noise caused by dark current determine the threshold of sensitivity and limit the ability of the charge-coupled array to record sources of radiation that are weak in terms of brightness. Recording of synchrotron radiation by a radiant intensity comparator based on a telescope with charge-coupled array is used to discriminate an individual electron on the orbit of an electronic storage ring. The least discrete decrease in the signal corresponds to a loss of an individual electron in the accelerator. This makes it possible to practically eliminate the component of the total standard deviation of the reproduction as a consequence of an error in the measurement of the number of particles in the reproduction of the spectral density unit of radiant intensity.

In the range of wavelengths 0.001–1.600 μm, a source of synchrotron radiation supports the reproduction of the spectral density of irradiance in the range 10^8–10^{14} W/(sr·m^3); the spectral density of a flux of radiation in the range 10^1–10^6 W/m; the spectral density of irradiance in the range 10^4–10^{10} W/m^3 with standard Type A uncertainty $u_A = (0.3–1.0)\cdot 10^{-2}$ and standard type B uncertainty $u_B = (0.1–0.7)\cdot 10^{-2}$; the spectral density of radiant intensity in the range 10^3–10^9 W/(sr·m) with standard type A uncertainty $u_A = (0.01–0.05)\cdot 10^{-2}$ and standard type B uncertainty $u_B = (0.015–0.03)\cdot 10^{-2}$; a flux of radiation in the range $1\cdot 10^{-6}$–$2\cdot 10^{-2}$ W with $u_A = (0.2–0.8)\cdot 10^{-2}$, $u_B = (0.1–0.35)\cdot 10^{-2}$; and radiant intensity in the range $1\cdot 10^{-3}$–$1\cdot 10^2$ W/sr with $u_A = (0.2–0.8)\cdot 10^{-2}$, $u_B = (0.1–0.35)\cdot 10^{-2}$.

The results of studies of the metrological characteristics of standards of the spectroradiometry of ultraviolet radiation and the results of international comparisons demonstrate a high level and reliability of reproduction and dissemination of the units corresponding to the modern level of the leading national metrological institutes.

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