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LABORATORY TECHNIQUES

A Facility for Studying Luminescence of Cryogenic Liquids Excited by β Particles and Positrons from ^{64}Cu Radionuclide

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Abstract—The design of a facility for studying luminescence of cryogenic liquids (nitrogen and helium) is described. Luminescence is excited by ~0.573-MeV β particles and 0.656-MeV positrons emitted by a radioactive source based on ^{64}Cu radionuclide, which is immersed in a liquid.

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As a high-energy charged particle passes through a dense material, it ionizes and excites atoms and molecules of the analyzed material along its track. After radiative decay (and recombination), a significant fraction of atoms and molecules appears to be concentrated at lower metastable levels, the transition from which into the ground state is forbidden by the selection rules. For example, for helium, these are the 2^1S and 2^3S levels, the energies of which are 20.61 and 19.82 eV, respectively; for nitrogen, this is the lower $A^3\Sigma_u^+$ state of a N_2 molecule with an energy of ~6.4 eV. As a result of binary, triple, and even quadruple collisions in a dense material, these long-lived excited particles form metastable polyatomic molecules (clusters). As was shown in [1, 2], such clusters may exist for tens of seconds. Information about the existence of helium and nitrogen clusters can be obtained from experimental investigations of luminescence in a material [3, 4] by measuring its molecular composition with a mass spectrometer [5–7] or examining the molecular structure of the liquid using the X-ray diffraction technique.

In this paper, we describe a facility for studying the luminescence spectra from cryogenic liquids excited by ~0.573-MeV β particles and 0.656-MeV positrons. The facility consists of an optical cryostat filled with a cryogenic liquid, a radioactive source of β particles, and positrons that is located directly in the liquid, an МДР-23 diffraction spectrometer that is a part of the СДЛ-2 facility, a cooled ФЭУ-100 photomultiplier tube (PMT) operating in a single-photon counting mode, a Ч3-34А frequency meter, and a computer.

The use of a cylindrically shaped radioactive source is the main feature of the facility. Such a source provides a means for obtaining higher flux densities of particles under investigation relative to a source of a standard shape (e.g., in the shape of flat disks). The source is made from a copper foil rolled up into a cylinder. The foil thickness (20 μm) is much smaller than the path length of emitted particles in the substance of the source; the cylinder radius (8 mm) exceeds the particle paths in a cryogenic liquid; and the cylinder height (50 mm) is limited by the absorption factor of luminescence light in the cryogenic liquid, the cryostat's vertical size, and the safety rules (the maximum source activity). In geometry such as this, all expected events will be localized inside the cylindrical source, which allows us to reduce the luminescence observation region.

The schematic layout of the optical cryostat and the recording module is shown in Fig. 1. Vessel *1* for the cryogenic liquid—liquid nitrogen—is made from a 35-mm-diameter 200-mm-long quartz tube with a flat window *2* welded to its bottom end. The quartz window is 3 mm thick. The vessel is filled with liquid nitrogen with a volume of ~100 cm^3, which is sufficient for taking continuous measurements over 2 h. To ensure thermal insulation, the quartz tube is enclosed in evacuated stainless steel shell *3* with a diameter of 72 mm.

Light emitted in the liquid is transmitted via windows *2* and *5* to the entrance slit of МДР-23 spectrometer *4* with the aid of controlled rotary mirror *6*. The mirror is located inside the heat-insulating housing and has regulating units for tuning its position. Quartz exit window *5* is attached to the side surface of the shell and is fixed in place by К-400 glue. Access to the regulating units of the rotary mirror is via hermetically sealed port *7*. Output pulses of PMT *8* are recorded by Ч3-34А frequency meter *9*, readings of which are saved to a file by computer *10*.

A small-sized incandescent lamp inserted into the cryostat in place of the radioactive source is used to align the recording system. After the mirror is aligned, the port is closed, the lamp is removed from the cryostat, and air is exhausted from the internal cavity of the shell. Afterward, nitrogen in supplied, β source *11* is set in the operating position, and the quartz tube is covered from the top with foam-plastic plug *12*. This is

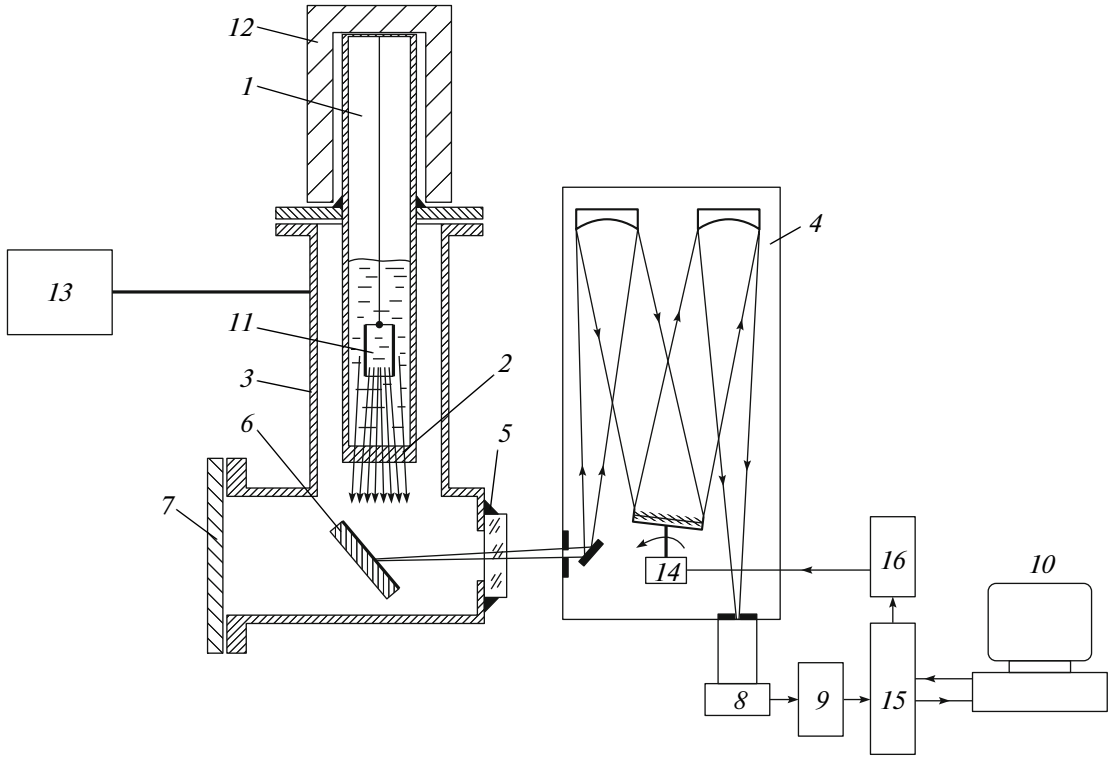

Fig. 1. Block diagram of the experimental facility for measuring luminescence spectra of liquid nitrogen: (*1*) vessel for cryogenic liquid, (*2*) window, (*3*) evacuated shell, (*4*) МДР-23 spectrometer, (*5*) quartz exit window (∅35 × 3 mm), (*6*) mirror, (*7*) port, (*8*) ФЭУ-100 PMT, (*9*) Ч3-34А frequency meter, (*10*) IBM PC computer, (*11*) ^{64}Cu source of β particles and positrons, (*12*) foam-plastic plug, (*13*) vacuum pump, (*14*) ШДР-711 step motor, (*15*) interface module, and (*16*) step motor control unit.

done to reduce the heat loss and prevent condensation of air components.

An optical cryostat for investigating luminescence in liquid helium has been designed, produced, and tested. There are three quartz windows in the bottom of the cryostat along its axis: a "cold" window in the helium vessel, which is sealed with an indium gasket; a window in the nitrogen screen, and a window in the outer housing. The last two windows are glued in by means of K-400 glue. The diameters of these windows are 16, 24, and 32 mm, respectively. A rod with the cylindrical β source is inserted in the cryostat along its axis. The lower edge of the cylindrical source is located at a distance of 1 mm from the "cold" window.

The cryostat is 530 mm high and 230 mm in diameter. The helium volume is 2.3 l. The helium consumption upon its initial filling with the preliminary cooling by liquid nitrogen over 2 h is 4 l and, upon inserting the rod with the source, 0.3 l. The time it takes for the liquid helium to evaporate to the level of the upper source's edge is 30 h. Pumping out helium vapor helps to lower the temperature to 1.5 K. The operating time at this temperature is 14 h.

The highest possible heat power released in the liquid helium by the β source is 2.5×10^{-3} W. This results in additional evaporation of helium at a rate of 3.6×10^{-3} l/h; in this case, the operating time of the cryostat decreases only slightly.

By contrast to the nitrogen cryostat, the mirror in the helium cryostat is enclosed in an untight light-proof housing, which is simultaneously used as a support for the cryostat. Access to the mirror for alignment is provided via the port.

The cryostat is also suitable for works with nitrogen in both liquid and solid states.

The cylindrical radioactive source (*11* in Fig. 1) of β particles and positrons on the basis of ^{64}Cu radionuclide is used to induce luminescence of a cryogenic liquid. The source is produced from a standard 20-μm-thick copper foil folded in a cylinder 16 mm in diameter and 50 mm in height, which is preliminarily irradiated in a nuclear reactor for a few hours. After exposure to thermal neutrons, the foil becomes radioactive and emits a flux of 0.573-MeV β particles and 0.656-MeV positrons with intensities ranging from 10^6 to 10^9 particles/(cm^2 s), depending on the exposure duration. In this case, the total power dissipated by the source in the cryogenic liquid ranges from 2.5×10^{-6} to 2.5×10^{-3} W. The half-life of the ^{64}Cu isotope $T_{1/2} =$ 12.8 h. The foil thickness (20 μm) is significantly smaller than the path length of an electron with an energy of ~0.5 MeV in copper, which is estimated to be

288 μm [8]. Therefore, self-absorption of β particles in the source can be ignored.

Naturally occurring copper is a mixture of two isotopes— ^{63}Cu (69.1%) and ^{65}Cu (30.9%). When a copper foil is irradiated with thermal neutrons, the following nuclear reactions proceed:

$$^{63}\mathrm{Cu} + n \longrightarrow {}^{64}\mathrm{Cu},\ \sigma(n, \gamma) = 4.5\ \mathrm{b}; \quad (1)$$

$$^{65}\mathrm{Cu} + n \longrightarrow {}^{66}\mathrm{Cu},\ \sigma(n, \gamma) = 2.2\ \mathrm{b}. \quad (2)$$

New copper isotopes ^{64}Cu and ^{66}Cu obtained thereby undergo decay with different half-lives according to the following schemes [9].

The main isotope:

$$^{64}\mathrm{Cu} \rightarrow {}^{64}\mathrm{Ni} + \beta^{+}(0.656\ \mathrm{MeV}) + \gamma(0.51\ \mathrm{MeV}),\ p = 19\%; \quad (3)$$

$$^{64}\mathrm{Cu} \rightarrow {}^{64}\mathrm{Zn} + \beta^{-}(0.573\ \mathrm{MeV}),\quad p = 38\%. \quad (4)$$

Half-life $T_{1/2}$ of reactions (3) and (4) is 12.8 h. The other 43% of ^{64}Cu decays are nonradiative and occur by capture of orbital electrons.

Secondary-product ^{66}Cu isotope decays as follows:

$$^{66}\mathrm{Cu} \rightarrow {}^{66}\mathrm{Zn} + \beta^{-}(1.65\ \mathrm{MeV}) + \gamma(1.05\ \mathrm{MeV}),\ p = 9\%; \quad (5)$$

$$^{66}\mathrm{Cu} \rightarrow {}^{66}\mathrm{Zn} + \beta^{-}(2.63\ \mathrm{MeV}),\quad p = 91\%. \quad (6)$$

Half-life $T_{1/2}$ of the secondary-product isotope for reactions (5) and (6) is 5.1 min.

The measurement time for one spectrum in the wavelength range of 200–800 nm is ≥10–20 min, depending on the selected scanning step and the data integration time. Therefore, we used only decay products of long-lived radionuclide ^{64}Cu to excite luminescence of cryogenic liquids. To get rid of minor decay products of ^{66}Cu, the source was held in air for ~3 h after its irradiation in the reactor before loading into the cryostat. This time was enough for complete decay of the ^{66}Cu radionuclide.

The spectroscopic system is controlled in an automatic mode with the aid of computer *10*. The following measurement parameters are specified: the beginning and end of the spectrum scanning range, the scanning step, and the data integration time at each spectral point. A spectrum is scanned by rotation of the diffraction grating, which is driven by ШДР-711 step motor *14*. A command for control module *16* of the step motor is outputted by the computer via interface unit *15*. Results of measurements are transmitted over the communication line via interface unit *15* to the personal computer and are saved to a file.

A ТРШ-2850 broadband incandescent lamp operating in the rated power supply mode (the filament current at a constant voltage of 6.4 V is 7.86 A, and color temperature of the lamp T_{color} = 2850 K) was used for energy calibration of the facility. The lamp was placed in front of the spectrometer with the cryostat being removed. Due to the high photodetector sensitivity, the lamp spectrum was measured with narrow slits limited by height by a round aperture 2 mm in diameter. The width of both the entrance and exit slits was 0.03 mm. The detection efficiency for photons incident on the entrance slit of the monochromator was calculated using the technique described in [10, 11]. Brightness $B(\lambda, T)$ [W/(cm^2 sr μm)] of the reference lamp, referred to 1 cm^2 of the emitting surface of the tungsten strip, unit solid angle, and unit wavelength range, is presented by the equation

$$B(\lambda, T) = \tau(\lambda)\varepsilon(\lambda, T)b(\lambda, T), \quad (7)$$

where $\tau(\lambda) = 0.92$ is the transmittance of the lamp's sapphire window over the whole wavelength range of visible light, λ [nm] is the radiation wavelength, T [K] is the absolute temperature of the tungsten strip, $\varepsilon(\lambda, T)$ is the emissivity factor of tungsten (the values of $\varepsilon(\lambda, T)$ were presented for different λ and T in [11]), and $b(\lambda, T)$ is the brightness of radiation from an absolutely black body from 1 cm^2 of the light source's surface in unit wavelength range and unit solid angle.

According to [10, 11], $b(\lambda, T)$ is

$$b(\lambda, T) = c_1[\exp(c_2/(\lambda T)) - 1]^{-1}\lambda^{-5}, \quad (8)$$

where $c_1 = 1.1909 \times 10^{-12}$ W cm^2 sr^{-1}, $c_2 = 1.438$ cm K.

The true temperature of the tungsten strip T was determined by the red–blue ratio (λ_1 = 470 nm, λ_2 = 655 nm) for color temperature T_{color} = 2850 K:

$$1/T - 1/T_{color} = \ln[\varepsilon(\lambda_1, T)/\varepsilon(\lambda_2, T)]/[c_2(1/\lambda_1 - 1/\lambda_2)]. \quad (9)$$

For our conditions, it appeared to be that $\varepsilon(\lambda_1, T) = 0.45$, $\varepsilon(\lambda_2, T) = 0.428$, and T = 2800 K.

Detection efficiency E_{eff} [count/photon] for photons emitted by the reference lamp and incident on the entrance slit of the monochromator was calculated according to the formula

$$E_{eff} = N_{strip}(\lambda)h\nu l^2 \times 1.6 \times 10^{-19}/[b(\lambda, T)\varepsilon(\lambda, T)d\delta S_{strip}S_{slit}], \quad (10)$$

where $N_{strip}(\lambda)$ [counts/s] is the measured counting rate at wavelength λ; $h\nu$ [eV] is the photon energy, l [cm] is the distance from the incandescent filament of the reference lamp to the entrance slit of the monochromator, 1.6×10^{-19} J/eV is the conversion coefficient; $b(\lambda, T)$ [W cm^{-2} sr^{-1} μm^{-1}] is the brightness of radiation of the absolute black body at temperature

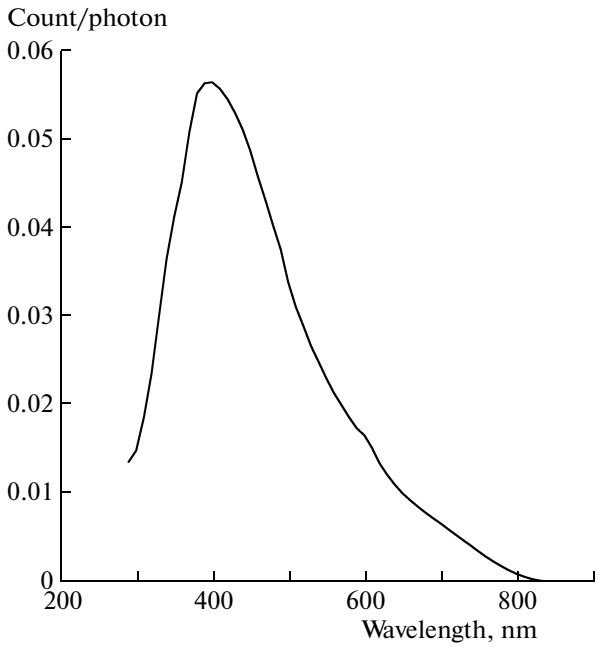

Fig. 2. Photon detection efficiency $E_{\text{eff}}(\lambda)$ of the МДР-23 monochromator with the ФЭУ-100 PMT.

$T = 2800$ K, $\varepsilon(\lambda, T)$ is the degree of grayness of tungsten at $T = 2800$ K, $d = 3.9 \times 10^{-5}$ μm/mm is the inverse linear dispersion of the monochromator, $\delta = 0.03$ mm is the exit slit width, $S_{\text{strp}} = 0.1 \times 0.7$ cm^2 is the area of the lamp's tungsten strip; and $S_{\text{slit}} = 0.03 \times 2.00$ mm^2 is the area of the monochromator's entrance slit.

The E_{eff} values calculated by Eq. (10) in the wavelength range of 300–850 nm for the МДР-23 monochromator with the ФЭУ-100 PMT are plotted in Fig. 2.

To estimate the light yield in the luminescent medium, one should know the number of photons produced inside the chamber per a 1 MeV-energy deposited in the substance under investigation. To do this, one must determine the total energy deposited by β particles from the source inside the chamber. The activity of the radioactive source was estimated, proceeding from the dimensions of the foil, its irradiation time in the core of the ИРТ reactor at the Moscow Engineering Physics Institute, and the neutron flux value. The calculated activity of the ^{64}Cu radionuclide in our cylindrical source exposed for 20 h to thermal

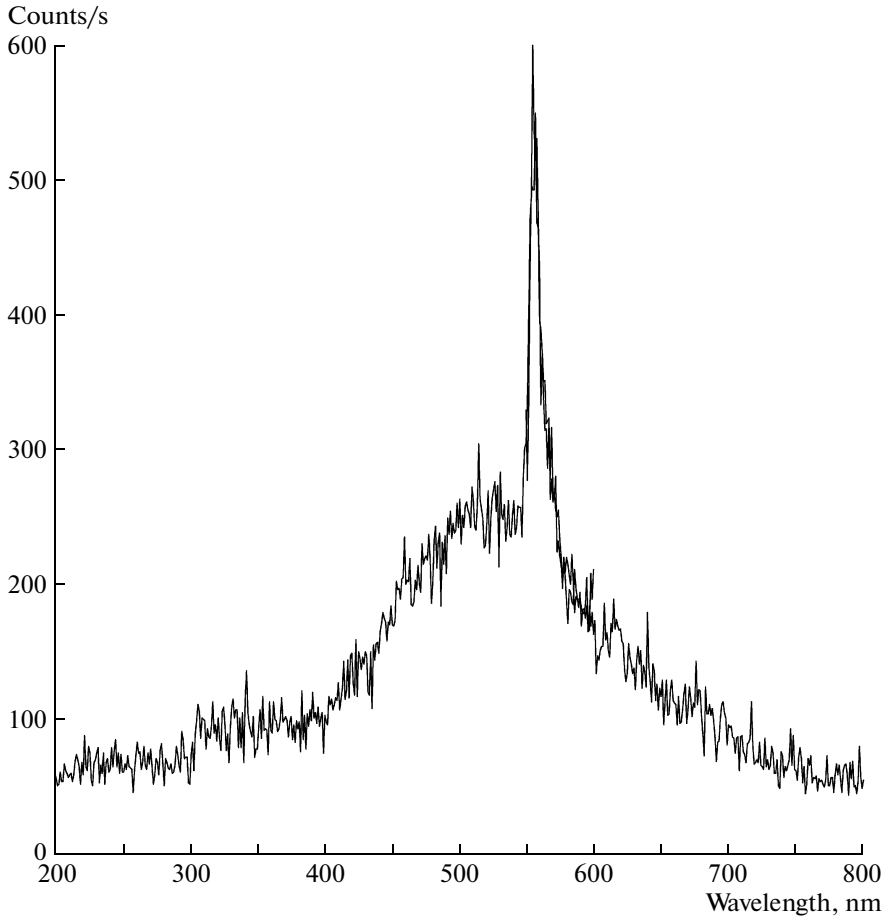

Fig. 3. Recorded luminescence spectrum of the liquid nitrogen sample excited by β particles and positrons of the ^{64}Cu isotopic source.

neutrons with a flux density of 10^{11} neutrons/(cm^2 s) was ~2×10^9 decays/s.

The luminescent region in the cryostat is a bulk extended light source with dimensions close to the source dimensions (∅16 × 50 mm). In the measurements, the cryostat was directly attached to the entrance slit of the monochromator (see Fig. 1). In such geometry, the photodetector viewed only a part of the cryostat's inner volume.

Thereafter, when processing the results of measurements of luminescence in the cryogenic liquid, we used a coefficient that took into account the incomplete light collection. For the given geometry, its value was 1.8. In view of the notes made, conversion efficiency K [photon/MeV] for the luminescent radiation of the cryogenic liquid excited by β particles of the ^{64}Cu source was calculated as follows:

$$K = N_{\text{meas}}(\lambda)4\pi L^2/(1.8E_{\text{dep}}E_{\text{eff}}(\lambda)S_{\text{slit}}), \qquad (11)$$

where $N_{\text{meas}}(\lambda)$ [counts/s] is the measured counting rate of photons with wavelength λ; $L = 400$ mm is the distance from the light source to the entrance slit, $E_{\text{dep}} = 5.25 \times 10^8$ MeV/s is the energy deposited by β particles from the source in the cryogenic liquid, $E_{\text{eff}}(\lambda)$ [counts/photon] is the detection efficiency for photons incident on the entrance slit of the monochromator, and S_{slit} [mm^2] is the area of the monochromator's entrance slit.

The luminescence intensity of the liquid nitrogen was measured in the wavelength range of 200–800 nm; the results of these measurements are shown in Fig. 3. Luminescence was observed as a wide continuum in the wavelength range of 390–760 nm with an intense molecular band of an unknown origin at 560 nm. The origin of this band may be associated with formation of nitrogen clusters under exposure to β particles. It is impossible to explain the origin of this band by, e.g., luminescence of molecular oxygen admixture in the liquid nitrogen for the following two reasons:

(i) the cryostat design rules out an ingress of condensed oxygen into the quartz sleeve with liquid nitrogen during spectrum measurement, and

(ii) the known O_2 molecular bands (the Hertzberg and Schumann–Runge bands, as well as forbidden atmospheric bands $^1\Delta_g - {}^3\Sigma_g^-$ and $^1\Sigma_g^+ - {}^3\Sigma_g^-$) differ radically in the wavelengths from the observed band at 560 nm.

The recorded spectrum also substantially differs from the luminescence spectrum of gaseous nitrogen excited by an electron beam, which is characterized by excitation of transitions $C^3\Pi_u - B^3\Pi_g$ of a N_2 molecule in the near UV region (337.0, 358.0, and 380.5 nm). Unambiguous identification of the measured spectrum calls for further experimental investigations of the time characteristics of induced luminescence.

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