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Synchrotron and undulator radiations and their applications in spectroscopy

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Synchrotron and undulator radiations currently enjoy wide application in all areas of science comprising radiation–matter interaction studies. In the theoretical and experimental investigation of synchrotron and undulator radiations, Moscow State University (MSU) initially had a close collaboration with the Lebedev Physical Institute of the Academy of Sciences. The discovery of synchrotron radiation was predicted by D D Ivanenko and I Ya Pomeranchuk in 1944. In their paper published in *Doklady AN SSSR* [1] (and, in parallel, in *The Physical Review* in the USA), D D Ivanenko and I Ya Pomeranchuk suggested that the reason for the limitation of accelerated electron energy in a betatron (a cyclic induction accelerator) lies with magneto-bremsstrahlung whose power is proportional to the fourth power of accelerated particle energy. In 1947, this radiation was discovered in the visible range in the General Electric 80-MeV synchrotron at the Brookhaven National Laboratory, USA. Hence, the magneto-bremsstrahlung of relativistic electrons (with velocities close to that of light) has come to known as synchrotron radiation. In 1948, D D Ivanenko and A A Sokolov [2] published a paper entitled "K teorii svetyashchegosya elektrona" ("On the theory of the luminous electron") also in Russian-language journal *Doklady AN SSSR*, where they reported their calculations of the corresponding angular and spectral characteristics of this radiation. The subsequent work of our theorists laid the foundations for the synchrotron radiation scientific school of Moscow State University [3–8]. As a result, the bibliographic list of the papers of MSU physicists concerned with the investigation of synchrotron and undulator radiations and their applications numbers now more than 1200, including several dozen Habilitation and PhD theses.

The first domestic experimental verifications of the theory of a luminous electron date back to 1956: the work by Yu M Ado and P A Cherenkov [9] on the energy distribution in the spectrum of the incoherent radiation of electrons orbiting a synchrotron, and the paper "Eksperimental'nye issledovaniya uglovogo raspredeleniya i polyarizatsii opticheskogo izlucheniya v sinkhrofazotrone" ("Experimental investigations of the angular distribution and polarization of optical radiation in a proton synchrotron") by F A Korolev, E M Akimov, V S Markov, and O F Kulikov [10] (Fig. 1), which appeared after the publication of the theoretical paper by A A Sokolov and I M Ternov on the polarization effects in the radiation of a luminous electron [11].

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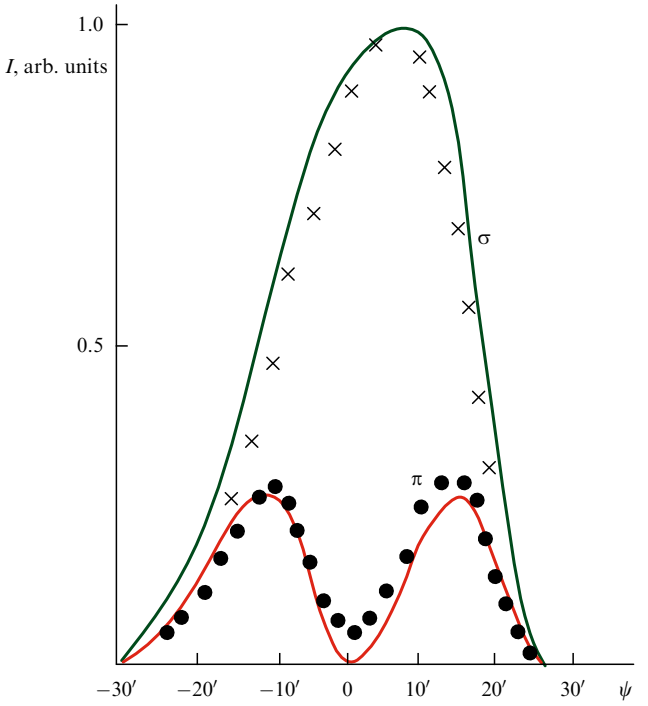

Figure 1. Comparison between the theoretical dependences (curves) of intensities I for σ and π linear polarized components on the angle ψ and the experimental ones (circles and crosses) (according to O F Kulikov) [10].

Experimental investigations of the properties of synchrotron radiation were subsequently carried out by MSU physicists (O F Kulikov, A S Yartsev, et al.) in collaboration with the Laboratory of High-Energy Physics of the Lebedev Physical Institute (LPI) of the USSR Academy of Sciences (Yu M Aleksandrov, M N Yakimenko, et al.). This research was encouraged by Academician D V Skobeltsyn, Director of LPI from 1951 to 1973. The first vacuum ultraviolet (VUV) spectroscopy channel built around the LPI S-60 680-MeV synchrotron was done under the auspices of D V Skobeltsyn back in 1967. More recently, research on the spectroscopy of solids has also been actively pursued in this country using other synchrotrons and storage rings, in particular, at the Institute of Nuclear Physics (INP), Siberian Branch of the USSR Academy of Sciences in Novosibirsk, and the Sibir'-1 storage ring at the I V Kurchatov Institute of Atomic Energy (IAE) in Moscow.

However, let us revert to the 1940s. Interestingly, even in 1948 A M Prokhorov was engaged in studies of synchrotron radiation for the purpose of obtaining coherent electromagnetic radiation in the centimeter and millimeter ranges of the spectrum. He executed a series of successful experiments to study the coherence properties of the radiation of relativistic electrons traveling in a uniform magnetic field. Prokhorov proved that synchrotron radiation may be employed as a source of coherent radiation in the centimeter range and determined the source characteristics and its power level. This research constituted the subject of A M Prokhorov's Habilitation thesis, which he successfully defended in 1951. It should be recalled, however, that the bulk of radiation power is confined in harder spectral regions — vacuum ultraviolet and X-ray ranges — and the main applications of synchrotron radiation emerge in precisely these regions.

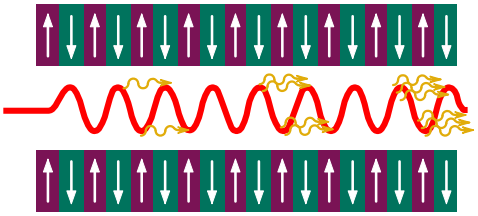

Figure 2. Simplest planar undulator scheme.

Synchrotron radiation (SR) and undulator radiation (UR) are common in nature, which is related to the emission of electromagnetic waves by accelerated relativistic charges [3, 4]. Nowadays, SR is a powerful universal instrument of research in various branches of science [5–7]. The technical progress and advancement of science of the past decades underlie the heightened interest in research on the stimulated emission of relativistic electrons (stimulated SR) (see, for instance, Refs [8, 12–16]). The features of stimulated SR appeared in electron motion along a helix were considered in Ref. [17], and stimulated SR in a focusing magnetic field of cyclic accelerators was comprehensively studied in paper [18]. The technological progress in the second half of the 20th century made it possible to make new SR sources — free-electron lasers (FELs), which comprise undulators. A significant difference between UR and SR is its high monochromaticity and high spectral density. The radiation of undulators with different field configurations has been adequately described in the literature (see, for instance, Refs [3–8, 12–19]). The simplest planar undulator is schematically depicted in Fig. 2. The first-ever observations of the radiation from an undulator built into the chamber of a cyclic accelerator were made in our country. In 1977, a team comprising LPI physicists supervised by P A Cherenkov and physicists from the MSU Faculty of Physics observed undulator radiation using the LPI's Pakhra synchrotron, which accelerated electrons to an energy of 1.2 GeV [20]. The team was able to take the first photographs of this new radiation type and study its spectral and angular characteristics, as well as the quasimonochromaticity effect [21].

The FELs made on the basis of UR sources have a significant distinction from ordinary lasers: the FEL operating frequency is not related to electron transitions to lower energy levels in a working medium, in contrast to ordinary laser frequencies. The function of the working medium in an FEL is in fact fulfilled by the electron beam traveling in the strong periodic magnetic field of an undulator, and the laser wavelength can be continuously tuned. The latter depends on the undulator parameters and the electron beam energy. The simplest version of an FEL is an undulator placed between two mirrors, one of which is semitransparent. The undulator, which produces incoherent radiation in a narrow frequency range about the frequency of the fundamental harmonic (or higher harmonics), is therefore placed into a resonator where laser radiation forms in its multiple transmissions through the undulator.

The new generation of SR–FEL sources operating in the X-ray region [22] invites new technical solutions, for instance, an FEL with self-amplified spontaneous emission (SASE). The SASE scheme implies the generation of coherent laser radiation in one undulator transmission. The SASE proceeds from the fact that electrons travel with a delay relative to their emitted SR which can catch up and interact with the electrons traveling ahead of it. In this way, the electromagnetic wave groups the electrons into separate bunches equally spaced at intervals equal to the radiation wavelength. As a result, several electrons begin to radiate nearly in phase. This effect becomes stronger with undulator length to give rise to denser electron bunches and pulsed coherent radiation.

In recent years, UR technology has been substantially improved not only from the standpoint of undulator magnets, but also from the standpoint of their configuration concepts. New findings of UR theory [23, 24] have shown how undulator schemes with a complex magnetic field configuration can enhance or weaken the emission of individual harmonics. Suppression of the hard UR component permits weakening its adverse effect on FEL mirrors, and the enhancement of specific harmonics may be employed in FELs with SASE and in devices exhibiting the high gain harmonic generation (HGHG), where use is made of a succession of undulators, each of which is tuned to a higher harmonic frequency of the previous one (see, for instance, Ref. [25]). In this connection, especially stringent requirements are imposed on the quality of the electron beam and the undulator design. All this calls for a rigorous and mathematically verified description of SR and UR properties with the inclusion of the features of radiation sources and their operating conditions; distortions of the trajectory of particle motion may have an adverse effect on the performance of the entire facility. Elaboration of UR theory with the application of generalized special functions enabled obtaining analytical expressions for the intensity and spectrum of UR with due regard for the constant component of the magnetic field [26–28], and, in particular, the terrestrial magnetic field. Therefore, the progress in UR theory makes it possible to produce devices with prescribed characteristics, preferred harmonics, and high quality for use in modern schemes intended for the generation of high-frequency coherent radiation in FELs with SASE, HGHG, etc.

Synchrotron and undulator radiations enjoy wide use in spectroscopy. Among important directions is the luminescent spectroscopy of solids excited by SR. The unique spectral characteristics and temporal structure of SR permit employing this radiation for the investigation of electronic relaxation in insulators with a wide forbidden band. Interpretation of these processes is critical, for instance, for understanding the scintillation efficiency of crystals. The method of investigating luminescence excitation spectra is convenient for studying energy transfer in these systems and investigating their electronic structure.

In general, luminescence excitation spectra may be subdivided into several spectral regions: direct excitation of the lower excited states of defects, defect ionization by photons with energies lower than the energy gap of the matrix, excitation in the Urbach tail of absorption by the matrix, exciton excitation, production of separated electron–hole pairs with a low energy, and production of high-energy electron–hole pairs with the subsequent collisional excitation/ionization of the matrix and defects.

Each of these regions is characterized by a different role of relaxation channels. In the absorption of photons of different energies in wide band gap insulators, diverse excitation types are produced in the crystal. The interrelation between these types of excitation and optical functions is depicted in Fig. 3 by the example of the dependence of the imaginary part of permittivity ε on the energy of absorbed photons, which directly reproduces the production efficiency of various

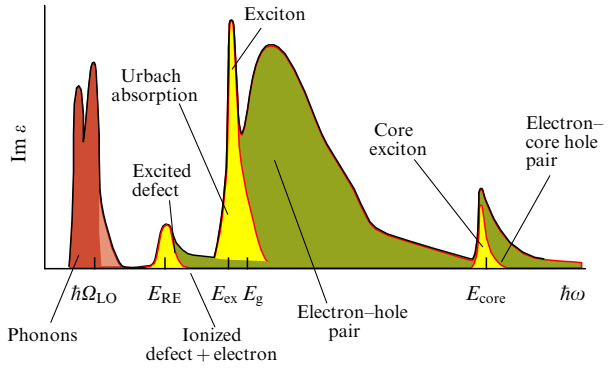

Figure 3. Imaginary part of the permittivity ε in a broad photon energy range and different processes investigated in the excitation of the luminescence of solids by synchrotron radiation: Ω_{LO} is the longitudinal optical phonon frequency, E_{RE} is the transition energy in a rare-earth activator, E_{ex} is the exciton energy, E_g is the forbidden gap width, and E_{core} is the core level energy.

primary excitations in crystals in relation to the energy of absorbed photons.

Table 1 shows the energy domains of exciting photons, the primary processes in solids corresponding to these energies, and those secondary processes of electronic excitation relaxation which may be studied using excitation by these photons. These processes will be considered below in greater detail by specific examples.

As a clear example, we outline the results of investigations of two types of BaF_2 luminescence excitation spectra (Fig. 4) in a broad spectral domain (from 5 eV to 1000 eV) [29]. This example is remarkable in that the relaxation processes of all types listed in Table 1 were investigated by one and the same technique in one substance.

Two types of luminescence are observed in BaF_2 — a relatively slow luminescence of self-trapped excitons (STEs) [its characteristic time at room temperature is about 600 ns (the solid curve in Fig. 4)], and a fast (with a 0.9-ns time) ultraviolet crossluminescence (CL) related to the filling of the upper core hole by an electron from the valence band (shown by circles in Fig. 4). It is readily seen from the initial portion of the spectrum that the excitation thresholds of these kinds of processes are different. For STE luminescence, this threshold coincides with the fundamental absorption threshold (about 10 eV), while for the core-valence transitions it coincides with the threshold of hole production in the outermost core band of barium (17 eV). It was precisely this difference in threshold energies that permitted LPI physicists at one time to unambiguously determine the nature of the fast ultraviolet emission in BaF_2 [30]. Clearly manifested in a higher-energy domain (from 25 to 90 eV) are the features of excitation spectra related to a near-surface loss. From the depth of dips in the excitation spectrum it is possible to determine the diffusion coefficient for elementary excitations in insulators. The excitation energy region close to 100 eV is of concern to us for the following reason: clearly manifested in it are the effects related to the production, by one photon, of several closely lying excitations either due to inelastic electron–electron scattering (for an energy below 100 eV) or due to a cascade of Auger processes following a 4d core excitation of barium (for an energy above 100 eV). These effects attendant upon a high local electronic excitation density manifest themselves both in a lowering of the luminescence yield and in kinetics acceleration due to mutual excitation quenching [31]. In BaF_2, there is no resonant interaction with core levels in a broad energy range from 280 to 680 eV. In this case, one might expect a linear increase in quantum yield with increasing photon energy (the number of low-energy electron–hole pairs resulting from a cascade of inelastic processes). However, noticeable departures from a linear shape are observed. These departures are attributed to the nonproportionality problem of scintillators' light yield, which has attracted considerable recent attention [32]. The highest attainable energy resolution of scintillators is substantially limited owing to this nonproportionality. The nonproportionality is primarily due to density effects which emerge in the tracks of ionizing particles. Finally, the investigation of relatively deep cores reveals clearly that the energy relaxation following the production of core holes on different ions proceeds along various paths. Usually, dips are observed in the excitation spectra when the photon energy passes through

Table 1. Energy relaxation processes in the excitation of crystals, which may be investigated using synchrotron radiation.

	Excitation region	Electronic excitation types	Relaxation processes
Visible light UV radiation	$h\nu \leqslant E_g$ $h\nu = 3-10$ eV	Excited and ionized states of defects and impurities. Self-trapped excitons	Emission of scintillators and phosphoruses. Intercenter energy transfer. Production of defects in the annihilation of excitons. Intracenter quenching
VUV radiation	$E_g \leqslant h\nu \leqslant (2-3)E_g$ $h\nu = 5-20$ eV	Electron–hole pairs with energies below the threshold energy for the production of secondary electronic excitations. Free excitons	Electron–phonon interaction resulting in thermalization and migration quenching due to separation of electron–hole pair components. Excitation diffusion. Excitation capture. Specific types of intraband and interband relaxation (for instance, crossluminescence)
Ultrasoft and soft X-rays	$(2-3)E_g \leqslant h\nu \leqslant (5-10)E_g$, $h\nu = 15-100$ eV	Hot excitations with energies exceeding the energy threshold for the production of secondary electronic excitations. Excitations of upper core levels	Inelastic electron–electron scattering and Auger processes resulting in cascade multiplication of electronic excitations
	$h\nu \geqslant (5-10)E_g$, $h\nu > 50$ eV	Core excitations	X-ray fluorescence. Auger processes
		Excited domains with several dozen excitations. Tracks of ionizing particles	Interaction involving a large number of excitations

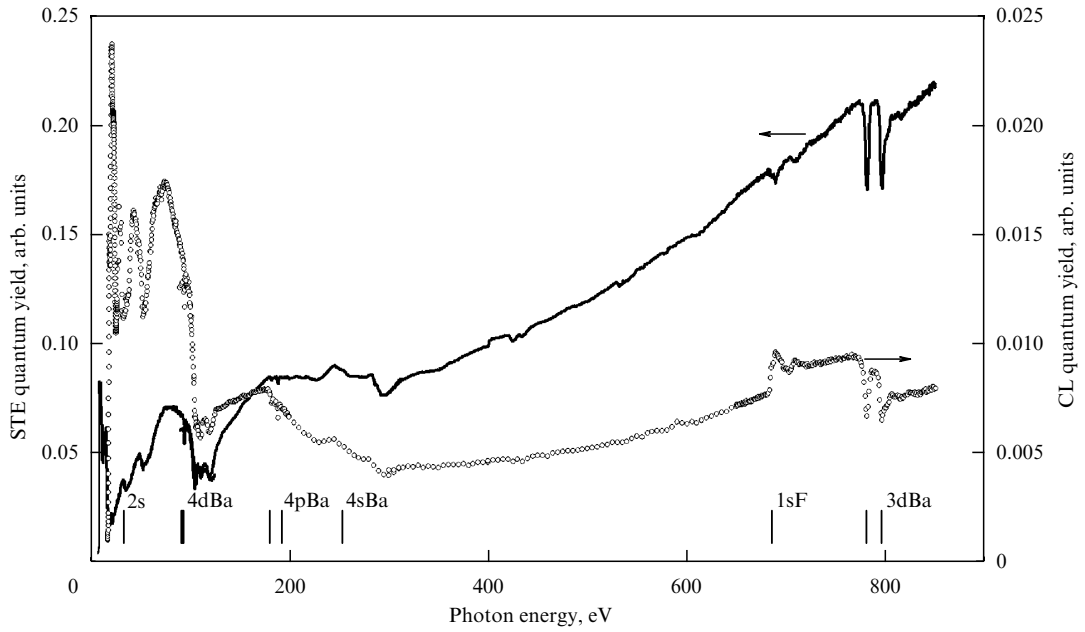

Figure 4. Excitation spectrum of two BaF_2 emission bands: of the emission of self-trapped excitons (STEs) (solid line), and of crossluminescence (CL) (core-valence transitions — circles) [29].

core energies (as is evident for low energies). However, a discontinuity is observed in the CL excitation spectrum in the 1sF domain, which is indicative of a new channel of 5p core hole production in Ba in the course of Auger relaxation [29].

Now let us consider several examples of the manifestation of these diverse processes in oxygen-containing wide band insulators. Such crystals are characterized by a rather broad valence band.

In the excitation by photons with an energy on the order of the fundamental absorption threshold, defect states are excited and ionized; then, for higher energies, processes emerge in the Urbach absorption domain, after which we enter the exciton domain of the spectrum. As the first example, we consider ytterbium-doped oxides of yttrium, lutetium, and scandium [33]. Observed in these crystals is luminescence with a charge transfer, whereat the excited state of an ytterbium center makes up a hole transferred from ytterbium to the nearest oxygen ions. The excitation spectra of these systems are clearly seen to divide into the following domains: the Urbach absorption domain, where defects are ionized; the exciton domain with direct excitation of the matrix, and the domain of production of separated electron–hole pairs. In particular, it is seen from the relationship between the fast and slow components of emission excited in the Urbach absorption domain that the slow component increases with an increase in the delocalization of holes (the closer to the edge of fundamental absorption, the more delocalized the state produced).

We now turn to the data on the excitation spectra of a scintillator which is familiar to everyone in the last ten years — lead tungstate (PWO) — for the application of this scintillator has played a critical role in the successful realization of CERN Large Hadron Collider detectors. Let us consider the processes proceeding in the domain of the onset of fundamental absorption (Fig. 5) [34]. The specific behavior of the excitation spectra of the fast (blue) emission of lead tungstate in relation to the temperature is a direct evidence that this emission is associated with the Urbach absorption edge of the lead exciton. This is caused by the

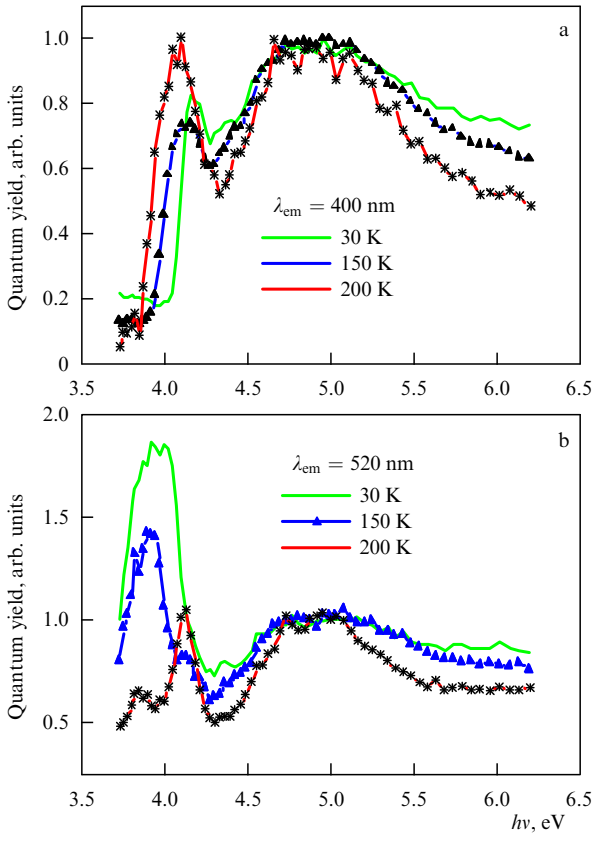

Figure 5. Excitation spectra of lead tungstate for the blue (a) and green (b) emission bands, which show the excitonic character of the fast (blue) emission and the recombination character of the green (a slower one) emission of this scintillator [34] (λ_{em} is the luminescence wavelength).

variation of the radiation fraction absorbed by the base matrix of the crystal under temperature variation, and testifies to the intrinsic nature of this emission. We will revert to these two emission bands more recently. We draw the

reader's attention to the temperature dependence of the excitation spectra of the blue emission (see Fig. 5) in the domain of production of separated electron–hole pairs: the dip deepens with increasing temperature. This effect is related to the following spectral domain, namely the production domain of separated electron–hole pairs. Depending on the kinetic energy of the primary electron–hole pair produced in the absorption in the domain of fundamental absorption, the fraction of those pairs varies which either are instantaneously coupled with each other to form an exciton-like state or, conversely, fly apart, which subsequently results in the emergence of slow luminescence components or in the radiationless recombination of excitations [34]. Since the recombination radius (the Onsager radius) depends strongly on the temperature, it is possible to trace the tendency of a variation of the spectral distribution along the channels with increasing temperature.

This effect is clearly seen not only in PWO excitation spectra, but also in many other spectra. In particular, reference can be made to the data on zinc tungstate [34]. This paper reports on the spectral dependence of the ratio between the excitation spectra for two different temperatures. This analysis technique has several virtues; in particular, it reduces the uncertainty in measurements of normalization spectra. Here, we see that the ratio between room-temperature and low-temperature spectra regularly lowers with increasing photon energy up to the threshold of electronic excitation multiplication (10 eV). The fact that this ratio comes to a nearly constant level, when the photon energy reaches a value of 13 eV, signifies that all primary excitations for these and higher energies decay and that the energy of these excitations turns out to be equal to about 6 eV on average.

Let us consider the domain of electronic excitation multiplication in greater detail. Electron–hole pairs with a sufficiently high energy are produced in this domain. The threshold of electronic excitation multiplication manifests itself most clearly in the combined investigation of luminescence and photoemission excitation spectra (see the example of MgO investigations [35] in Fig. 6). The sharp rise in luminescence in the excitation spectra for an energy of 19.5 eV may be ascribed not only to the multiplication, but also to a lowering of absorption and reflection coefficients; however, the behavior of the photoemission excitation spectrum suggests that precisely the multiplication threshold is observed at this energy.

The quantum yield, which was considered by the example of tungstates luminescence and which lowers with increasing photon energy, does not represent the only type of excitation spectra. When luminescence is related to recombination on defects, another type of quantum yield may also be observed. Different examples of the excitation spectra of this type are presented for the excitation of intrinsic luminescence of $CaWO_4$ [36] and CeF_3 [37], and activator luminescence of $CaSO_4:Sm$ [38].

Several words are in order about the role of core excitations. In the foregoing, we mentioned core excitations in BaF_2. Spassky et al. [39] drew attention to the fact that the investigation of optical functions in the domain of core excitons permits coming to conclusions about the character of the lower states in the conduction band. The existence of a sharp structure in the domain of cation excitons suggests that cation states for lead and barium in molybdates make a significant contribution to the bottom of the conduction

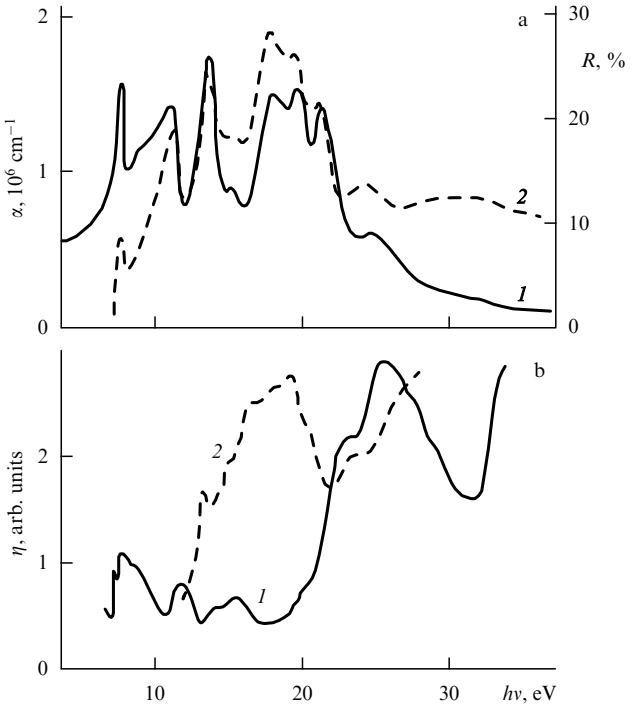

Figure 6. (a) Reflection (curve *1*) (R is the reflection coefficient) and absorption (curve *2*) (α is the absorption coefficient) spectra of an MgO-Al single crystal. (b) Luminescence (curve *1*) and photoemission (curve *2*) (η is the quantum yield of luminescence or photoemission) excitation spectra of an MgO-Al single crystal [35].

band, while the energy levels of strontium and barium in this family lie deep in the conduction band.

An example of spectral-kinetic investigations of cerium emission in more complex crystals was given in a report by Vasil'ev and Bel'skii made to the recent International Conference on Luminescent Detectors and Transformers of Ionizing Radiation (LUMDETR-2012) [40]. Different processes show their worth in these decay kinetics selectively excited at different excitation energies: direct excitation of cerium with monoexponential kinetics, ionization of cerium, dipole–dipole transfer from excited lutetium ions to cerium with subsequent ionization of cerium, change in the kinetics in the course of transfer from an exciton (Lu + F), transfer from electron–hole pairs, and transfer with quenching and ionization in the excitation of separated electron–hole pairs. Mathematical processing of kinetic curve shapes permits determining important parameters of energy transfer, in particular, the dipole–dipole interaction radii, the radii of flying apart of electron–hole pair components in its ionization, etc.

We now turn to a description of investigations of another practically important effect — the resistance of scintillators to radiation damage. Synchrotron radiation turned out to be a convenient tool for studying this problem as well [41]. Let us discuss the main effects related to radiation damage. These effects may be subdivided into reversible and irreversible. Reversible effects are primarily reduced to the production of short-lived defects (defect pairs) or to changes in the charge state of defects with deep energy levels. We will mostly consider precisely the latter case. Irreversible damage consists primarily in the production of stable pairs of elementary defects and conglomerates of defects. The unique spectral properties and temporal structure of SR, as well as its high

intensity, permit using this kind of excitation for the investigation of defects and their production processes in crystals with a wide forbidden band, in particular, in scintillators.

The technique of investigating the mechanisms of luminescence excitation is highly convenient for exploring the energy transfer in defect systems in different matrices, the energy structure of insulators, and their resistance to radiation damage.

Let us consider how radiation effects may manifest themselves in investigations based on the luminescent spectroscopy technique. These are mostly the changes of emission spectra under dose variations, which reveal themselves most often in the emergence of additional excitation bands. Next, these show up as changes in emission decay kinetics with increasing dose (radiation effects make themselves evident in the sharpening of the initial decay stage and the emergence of a slow emission component).

Changes in energy transfer are mostly manifested as changes in the ratio between the contribution probabilities from different relaxation channels in insulators.

What is the convenience of using VUV and X-ray SRs in the investigation of radiation damage? VUV and X-ray photons produce the same electronic excitations (electron–hole pairs, excitons, core-level holes, initial stages of defect production) as high-energy ionizing particles (because in the course of relaxation of ionizing particle tracks so-called delta electrons with energies from 20 to 10,000 eV emerge). Furthermore, the absorption coefficient in this spectral domain is so high that the photons are absorbed in a very thin layer and the dose accumulated in this layer is quite significant for moderate photon fluxes.

In summary, we notice that vacuum ultraviolet and X-ray synchrotron radiations permit one not only to examine the basic mechanisms of electronic relaxation and energy transfer in wide band crystals, but also to investigate the resistance of scintillators to radiation damage.

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The Petersburg period in the life and scientific work of D V Skobeltsyn

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They are not born to be outstanding scientists — they become such to the best of their research abilities and especially so due to their persevering labor. The success of such a formation is largely due to the foundation laid in a person in his green years. The foundation is determined both by biological genes (from one's parents) and by 'social genes' — from the family

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