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Losses of fast scintillation light
Ztráty rychlého scintilačního světla

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Summary

Scintillating materials are used in a variety of applications imposing specific requirements on their performance. A number of applications requires fast response of materials making any losses of fast scintillation light detrimental. Such losses are due to the presence of trapping centers localized in the material band gap. We address the processes contributing to the losses of fast scintillation light, focusing, in particular, on thermal ionization of the activator's (luminescent center) excited state and quantum tunneling between the activator and the trap. We present both experimental and theoretical studies of these processes in currently investigated scintillating materials.

Souhrn

Scintilační materiály jsou využitelné v široké škále aplikací, které kladou specifické požadavky na jejich technické parametry. Mnoho aplikací vyžaduje zejména rychlou odezvu materiálu, pro kterou jsou jakékoliv ztráty rychlého scintilačního světla velmi nepříznivé. K takovým ztrátám dochází v důsledku zýchytných stavů (pastí) lokalizovaných v zakázaném pásu materiálu. Popíšeme procesy, které přispívají ke ztrátám rychlého scintilačního světla se zaměřením zejména na termální ionizaci excitovaného stavu aktivátoru (luminiscenčního centra) a kvantového tunelování mezi aktivátorem a pastí. Ukážeme jak experimentální, tak teoretické studie zmíněných jevů v současně zkoumaných scintilačních materiálech.

Keywords

Scintillator, ionizing radiation, thermal ionization, tunneling

Klíčová slova

Scintilátor, ionizující záření, termální ionizace, tunelování

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1 Introduction

A scintillator is a material that converts the absorbed ionizing energy of a high energy photon (X- ray or γ -ray), an accelerated charged particle or neutron into a number of ultraviolet to visible (UV/VIS) photons. The scintillation detector consists of scintillating material coupled to a photodetector (photomultiplier tube or photodiode). The latter transforms the created UV/VIS photons into an electrical pulse [1,2]. Suitable scintillating materials are found within the category of wide band-gap semiconductors or dielectrics.

Scintillation is the process of luminescence excited by ionizing radiation. The scintillation mechanism and efficiency criteria were already addressed in [3], with later refinement in [4]. The scintillation mechanism can be divided into three consecutive processes: *conversion, transport and luminescence*; see Fig. 1.

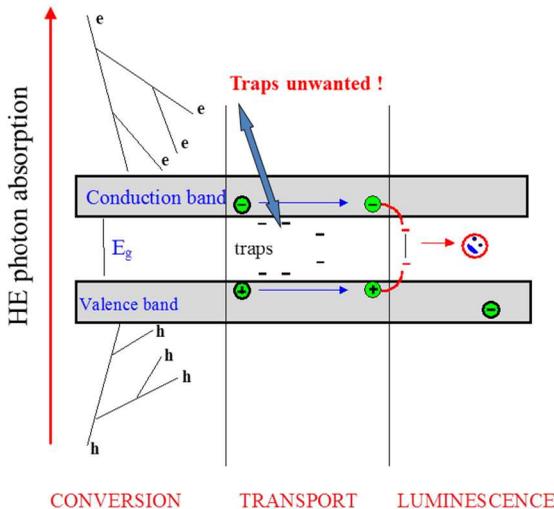


Fig. 1. Scheme of scintillation mechanism in a solid state crystalline material. After [2].

Depending on the particle energy, its initial multi-step interaction with the scintillator host lattice progresses predominantly *via* the photoelectric effect, Compton scattering and pair production. The hot electrons and deep holes that are created are gradually thermalized at the conduction and valence band edges, respectively. The conversion process typically lasts a few picoseconds. In the transport process the separated electrons and holes migrate through the host lattice to reach the emission centers. During their migration charge carriers can be (repeatedly) trapped or even nonradiatively recombine at trapping levels occurring in the host band gap. Due to the trapping process the delivery of charge carriers to luminescent centers can be substantially delayed. Therefore, the presence of localized energy states in the host band gap acting as trapping centers may considerably deteriorate scintillator performance. During the final stage, the trapping and radiative recombination of the electron and hole at the luminescent center generates the desired luminescence.

In the last two decades there has been vivid ongoing activity in the scintillator research field to meet the demands of a variety of applications requiring detection of ionizing radiation. The fields of interest include high energy physics [5,6], imaging systems for medical diagnosis [7], medical therapy [8], homeland security [9] and others. Different applications put different requirements on the specific parameters characterizing the scintillator material performance. In a number of applications, fast response of the material becomes crucial.

New scintillating materials currently reported are predominantly based on Ce^{3+} and Pr^{3+} activated materials, due to the short decay time (typically 10-60 ns) and high quantum efficiency of the $5d \rightarrow 4f$ radiative transitions of these centers at room temperature [10].

Prompt or fast scintillation light is due to the recombination of charge carriers that were – after thermalization – “immediately” delivered to the luminescence center, i.e. those not being trapped. As mentioned above, trapping at localized states causes a delayed delivery to the luminescence centers and generation of the slow scintillation light on the expense of the fast light. Besides trapping during the transport process, the trapping states may also contribute to

the slow light after delivery of charge carriers to luminescence centers. This would be due to processes of thermal ionization of the luminescence center excited state and tunneling between the luminescence centers and (nearby) traps.

2 Thermal ionization

Conversion of excitation energy into mobile charges can occur as a result of thermally stimulated ionization of the activator ions excited state. Studying this process helps with the placement of the interacting energy levels within the band gap of the host. Thermally stimulated ionization of impurity ions in ionic hosts can be studied via the “contact” method, namely classical photoconductivity measurements using blocking electrodes [11]. This method is limited to the study of bulk crystalline materials. “Contactless” methods, such as the microwave resonator cavity technique [12], or purely optical techniques involving thermally stimulated luminescence (TSL) excitation spectroscopy [13] TSL after UV excitation [14] or *delayed recombination decay* measurements can also be used for microcrystalline or nanocrystalline powders.

2.1 Delayed recombination decay technique

The principle of the method was first described and implemented in [15]. The activator center is selectively excited into its own absorption band. When, after the photoluminescence (PL) excitation, the excited state of the luminescent center is thermally ionized, charge carriers that do not decay promptly can migrate through the conduction (or valence) band. During that process they can be trapped and therefore delayed in their recapture at the luminescent center. Escape from a trap is often thermally assisted and if the charge carrier reaches the luminescent center, its time spent in the trap will cause it to be part of the observed slow tail of the PL decay. Consequently, monitoring the temperature dependence of the slow tail of the PL decay, or the so called *delayed recombination decay*, supplies information about the thermal ionization of the activator’s excited state.

The delayed recombination signal is monitored by conventional equipment for measurements of slow PL decay on the micro-millisecond time scale. The sample is irradiated into the absorption band of an activator by an intense low-repetition-rate pulsed light source (e.g., 10 Hz). The luminescence decay is scanned until the arrival of the next excitation pulse. The system is set so that each excitation pulse (and corresponding measurement window opening) occurs immediately after the previous one. The scheme of the measurement sequence is shown in Fig. 2. The accumulation time for each measurement is typically about 10 minutes. The decay is scaled within about 10-50 ms (light blue window in Fig. 2). The decay curve is then integrated after the background is subtracted and the first few channels that may contain the prompt nanosecond decay component are excluded. The integrated decay curve provides the overall intensity of the delayed recombination processes at an activator. The measurement is performed at different temperatures starting from the highest one. From the plot of temperature dependence of the delayed recombination intensity the thermal ionization of the activator can be assessed (see the next subsection). The technique was recently used for luminescence centers in a variety of materials, see, for example [16,17] to provide more insight into the processes occurring in the excited state of the luminescence center.

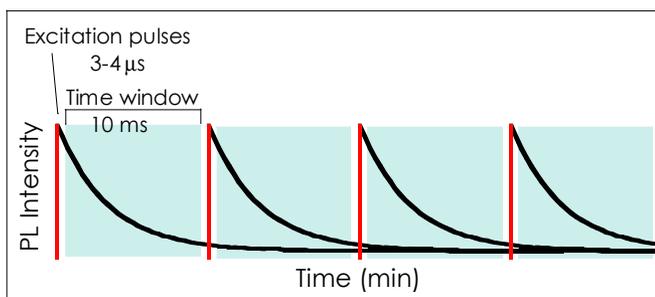


Fig. 2. The scheme of the delayed recombination measurement.

2.2 The model of temperature dependence of delayed recombination decay intensity

The theoretical model considering the role of traps in shaping the temperature dependence of the delayed recombination intensity was first proposed for the case of discrete traps [14] and then generalized for any trap distribution function [18].

The intensity of delayed recombination light is proportional to the fraction of thermally ionized charge carriers $N_I(T)$ that can be expressed as:

$$N_I(T) = w e^{-\frac{E_{ion}}{kT}}, \quad (1)$$

where T is the excitation temperature, w is a dimensionless pre-exponential factor, E_{ion} is the thermal ionization energy (the energy separation between the excited state of the luminescent center and the bottom of the conduction band) and k is Boltzmann's constant. However, not all of the thermally ionized carriers will contribute to the overall delayed recombination signal. In particular, only those that get trapped and subsequently released from the traps (with a particular trap depth) whose detrapping time falls into the time interval of delayed recombination measurement will contribute (see above). In general, the number of traps with a certain trap depth can be determined from the distribution function of traps. Thus the intensity of delayed recombination decay can be expressed as:

$$I_{DR}(T) = w e^{-\frac{E_{ion}}{kT}} \int_0^{\infty} dE p(E) \int_{t_b}^{t_e} dt \frac{1}{\tau(E)} e^{-\frac{t}{\tau(E)}}, \quad (2)$$

where $p(E)$ is the distribution function of traps with trap depth E and $\tau(E)$ is the detrapping time of the trap with depth E , t_b and t_e are times corresponding to the beginning and end of the delayed-recombination-decay signal recording, respectively. In the case of discrete traps, as in the crystalline hosts, formula (2) reduces to:

$$I_{DR}(T) = w e^{-\frac{E_{ion}}{kT}} \sum_{k=1}^n A_k \left[e^{-\frac{t_b}{\tau(E_k)}} - e^{-\frac{t_e}{\tau(E_k)}} \right], \quad (3)$$

with A_k being the amplitude of the trap with depth E_k and n the number of traps of various kinds participating in the process. The amplitude

A_k is proportional to the number of traps of the same kind contributing to the delayed recombination signal. The number of parameters entering formula (3) is relatively high, however, most of them namely the trap depths E and detrapping times, can be determined from an independent experiment using the TSL technique. The model was tested on a Pr^{3+} doped $\text{Lu}_2\text{Si}_2\text{O}_7$ (LPS) single crystal [14], Ce^{3+} -doped SrHfO_3 microcrystalline powder [18] and $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Eu}$ powder [P19], see Figs. 3-5.

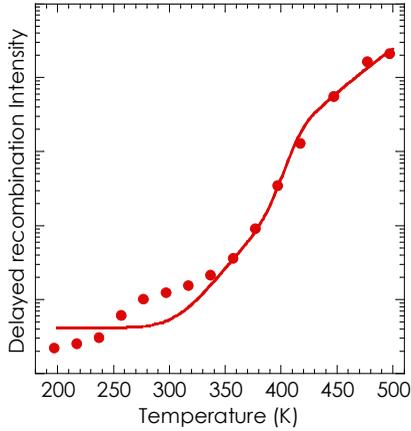


Fig. 3. Temperature dependence of the delayed recombination light emitted by LPS:Pr. The solid circles are experimental data, the line is the numerical fit of formula (3) to the data with $E_{\text{ion}} = 0.54$ eV (see also [14]).

3 Quantum tunneling

Tunneling can significantly affect the dynamics of luminescence center excited states and become a source of fast scintillation light losses. Delayed recombination decay measurement appears to be suitable to detect the presence of quantum tunneling as well.

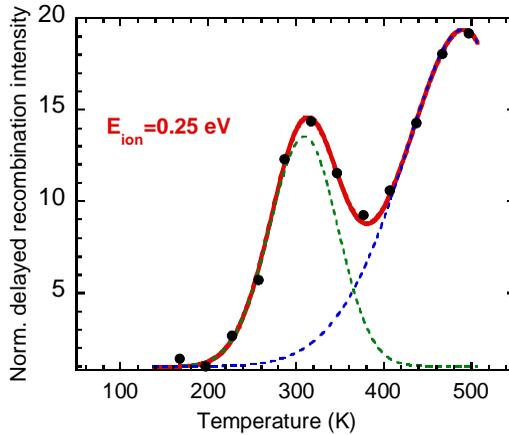


Fig. 4. Normalized (to minimum) delayed recombination intensity of SrHfO₃:Ce³⁺ as a function of temperature. Solid circles are experimental data. Solid line is the fit of Eq. (3) to the data, dashed lines are contributions of each single trap to the overall signal (see also [18]).

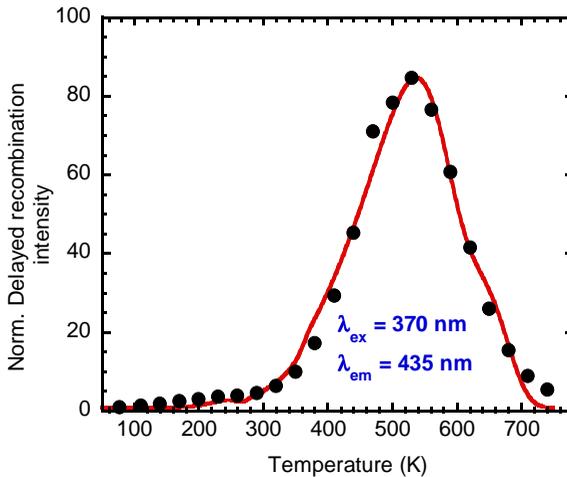


Fig. 5. Delayed recombination decay intensity of Y₃Al₅O₁₂:Eu as a function of temperature. Solid circles are experimental data. Solid curve is the fit of function (3) (in the text) to the data (see also [P19]).

The low temperature limit of delayed recombination decay intensity given by formula (2) is zero. Therefore, at low enough temperatures one would not expect to obtain a detectable delayed recombination signal due to thermal ionization of the luminescence center. However, this is not the case [18]. There is a low temperature contribution to the signal that is *not* due to thermal ionization of the luminescence center. A good candidate to explain the phenomenon appears to be quantum tunneling between the recombination center and a nearby trap(s). The idea has been pursued both experimentally and theoretically.

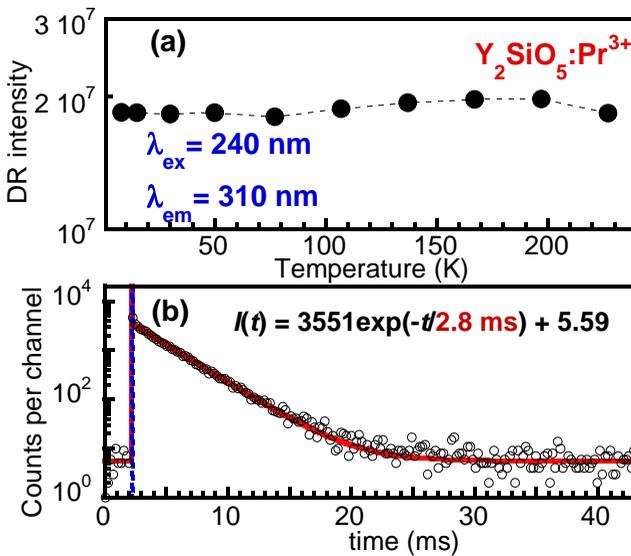


Fig. 6. In (a) is shown delayed recombination intensity of $Y_2SiO_5:Pr^{3+}$ as a function of temperature. In (b) is shown delayed recombination decay at 8 K. Empty circles are experimental data, solid line is the fit of function $I(t)$, given in the figure, to the data (see also [21]).

Since quantum tunneling is independent of temperature, one expects detectable signal at fairly low temperatures which should stay constant within certain temperature range. An extensive experimental support was obtained by several studies. The first support was

obtained for LPS:Pr single crystal [20], later for various complex oxide scintillating crystals grown by different techniques and doped with different activators [21], variety of scintillating Ce^{3+} and Pr^{3+} - doped simple garnets [22]. An example in Fig. 6 shows the delayed recombination intensity of $\text{Y}_2\text{SiO}_5:\text{Pr}^{3+}$ staying constant in the temperature interval 8-220 K. The delayed recombination decay at 8 K is approximated by a single exponential and represents a nice example of quantum tunneling between the activator and a single trap (the trap occurring at a single distance from the activator center.)

3.1 Models of quantum tunneling between the trap and recombination center

Delayed recombination decay measurements provide the decay times of the slow light on the order of microseconds to milliseconds. This corresponds to the decay slowdown by several orders of magnitude compared to prompt decay times of an activator. To address theoretically, whether quantum effects could be responsible for such slowdown, a simple one-dimensional model has been set up [P20]. A scenario of the underlying process is reflected by the scheme in Fig. 7a. The electron tunnels from the activator into a nearby trap. In principle the trap energy is *nearly* resonant with that of the excited activator state (reflected by a small energy separation $\Delta\epsilon$). The one-dimensional model is of the form $H=p^2/2m + V(x)$, with m the electron mass and $V(x)$ a potential:

$$V(x) = -\beta\delta(x) - V_0\theta(\ell - |x|) - \alpha\delta(x - \ell - \lambda), \quad (4)$$

with V_0 , α , β , λ being positive constants. The form of potential together with the definition of the parameters is shown in Fig. 7b. The parameters determine the trap energy, the ground and excited states of the activator and the distance between trap and activator. The time-independent Schrödinger equation is solved numerically by discretizing on the line. Fig. 8 presents the wave functions calculated for parameter values representing real systems and decay time delayed by six orders of magnitude. A presence of the trap wave function in the activator region supports the

hypothesis that quantum tunneling is responsible for the low temperature delayed recombination decay.

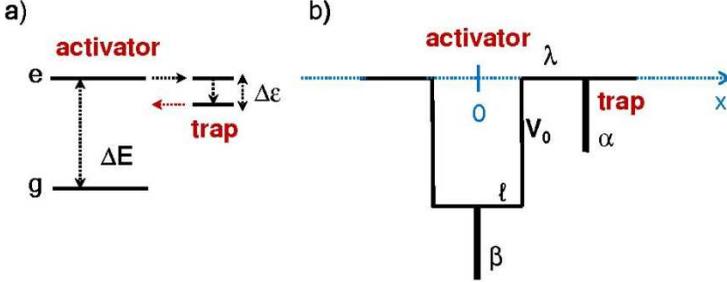


Fig. 7. (a) Simple scheme for a tunneling process between the activator, with the ground state g and excited state e , and the trap located nearby. The energy separation between the ground and excited state of the activator is ΔE while that of the excited state of the activator and the trap state is $\Delta \epsilon$. (b) The one-dimensional potential used in numerical calculations (see the text). The activator is placed at the origin (0) and V_0, ℓ, λ are the depth and half-width of the square as well as the strength of delta function representing the region of attraction of the activator, while α is the strength of delta function representing the region of attraction of the trap; $\lambda + \ell$ is thus the distance of the trap from the center of the activator. See also [20].

To allow a more stringent test of the tunneling hypothesis we set up a two dimensional model [23]. Both the trap and the ‘activator’ are taken to be isotropic, Gaussian wells. Specifically, the potential has the following form:

$$V(\rho, z) = -\alpha e^{-(\rho^2 + z^2)/r_A^2} + \beta/2 (\rho^2 + z^2) - \gamma e^{-(\rho^2 + (z-\lambda)^2)/r_T^2}, \quad (5)$$

where ρ is the 2D polar coordinate (along the line connecting the centers of the trap and activator) and z the orthogonal coordinate whose origin is the activator. The quantities α and γ are the depths of the activator and trap potential wells, respectively, β models the

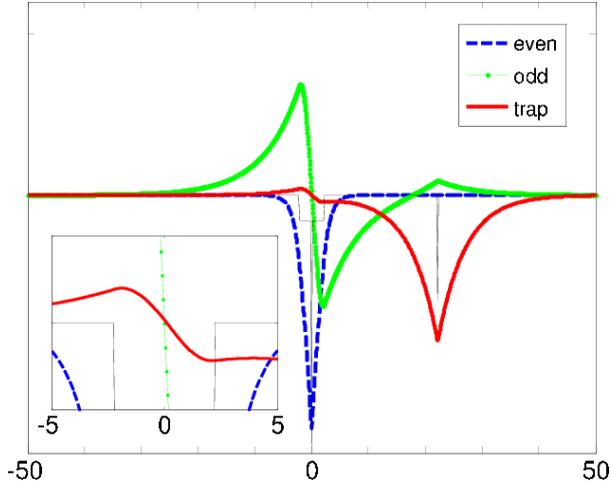


Fig. 8. Wave functions. A rescaled version of the potential is also shown. The solid (red) is the trap wave function, the dashed (blue) the even state (activator's ground state) and the dotted (green), the odd (activator's excited state). In the inset is the detail of the trap wave function in the region of the activator (see also [20]).

centrifugal potential and r_A and r_T are effective activator and trap radii, respectively.

Solution of the Schrödinger equation was achieved using the program 'WavePacket' [24]. In Fig. 9 we present an example of the trap wave function obtained from our numerical calculation. The right hand panel illustrates the presence of the trap wave function in the activator region.

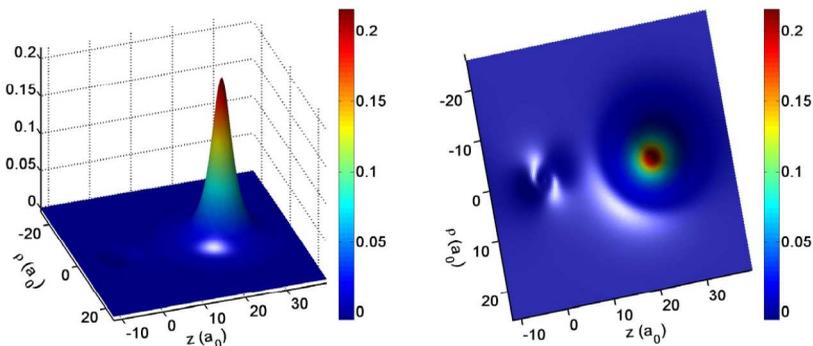


Fig. 9. Example of a calculated trap wave function shown from two different perspectives. The trap-activator distance λ is 23 Bohr radii (about 12 Å). The view in the second panel better displays the portion of the wave function in the area of the activator. The small asymmetry that can be seen as white spots on the dark background is an artifact of the solution method (see also [P23]).

4 General discussion and conclusion

An increasing number of applications of luminescent and scintillating materials in many fields of modern life imposes requirements on their performance. To meet the application-specific demands there is a continuous search for either new materials or ways to improve the existing ones. This task can only be handled by intense study of material characteristics and understanding the underlying mechanism of luminescence and scintillation as well as processes affecting light production. Indispensable tools that helps throughout are the methods of optical spectroscopy. They can, in particular, effectively monitor the excited state of the luminescence center, a feature that has a crucial role in assessing the scintillating material performance. Routine use of classical methods can be supplemented by novel techniques employing standard instruments in a specific inventive way.

A number of scintillating material applications requires the fast response of the material, i.e. production of fast light. Therefore, any losses of fast light are highly undesirable. Such losses originate due to the presence of traps in the material band gap. We addressed two

processes that can contribute to fast scintillation light losses. These are thermal ionization of the activator's excited state as a temperature *dependent* process and quantum tunneling between the activator and nearby trap(s), which is temperature *independent*.

We presented a delayed recombination decay technique, that can help to study both these processes. There are other methods available. However, the delayed recombination decay measurements are applicable to both crystalline and powder materials. When combined with TSL measurements the data obtained allow determination of the thermal ionization energy and of the position of the activator's excited state within the host's forbidden gap. The technique also makes it possible to assess the presence of quantum tunneling. The combination of the delayed recombination data with the data from independent experiments can get handle on the trap-activator distances. In this context the technique helps to understand the origin of temperature independent losses of fast scintillation light in scintillating materials. Although temperature independent losses are somewhat less intuitive than those that re temperature dependent, they do not seem to be infrequent. It is, therefore, of high importance that they be addressed and studied, for the ultimate goal would be to prevent them.

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5 Curriculum vitae

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Education

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Work experience

- **since 1996** - junior scientist in the Institute of Physics, Czech Academy of Sciences, Prague (CSc.)
- **since 2000** - senior scientist in the Institute of Physics, Czech Academy of Sciences, Prague (CSc.)
- **since 2004** - leading senior scientist in the Institute of Physics, Czech Academy of Sciences, Prague (CSc.)

Professional research interests in last 5 years:

Physics of scintillation and luminescent materials, study of luminescence and scintillation mechanisms, characteristics of new material systems in the form of bulk single crystals and ceramics, and nanopowders. Prizes and awards:

- "Prize of Otto Wichterle" of the Academy of Sciences of the Czech Republic (2002)

- The Prize of the Academy of Sciences of the Czech Republic for outstanding scientific results, 2003: „PbWO₄ scintillator for high energy physics. Physical description and material optimisation“.

Obtained grants/ domestic:

- "Optical characteristics of Jahn-Teller systems with an O_h symmetry", identif. code IAB1010901, Grant agency: AVČR, 1999-2000, budget: 0.86 mil. Kč, role: principal investigator
- joint NSF USA--ME CR project: "Coherent and incoherent decay of the doped alkali halide crystals" “, identif. code ME 382, Agency: ME, 2000-2002, budget: 4.85 mil. Kč, role: principal investigator
- "Nanostructures in dielectric crystal matrices: theory and experiment" identif. code IAA1010210, Grant agency: AVČR, 2002-2004, budget: 3.4 mil. Kč, role: principal investigator
- joint NSF USA-ME CR project: "Anomalous decay, slow relaxation and soliton creation in alkali halide crystals" identif. code ME 587, Grant agency: ME, 2002-2004, budget: 3.54 mil. Kč, role: principal investigator
- joint NSF USA--ME CR project: "Slow lattice relaxation and discrete breathers", identif. code ME 903, Grant agency: ME, 2007-2009 budget:1.2 mil. Kč, role: principal investigator
- joint Japan-ME CR project: "Development of novel scintillating materials for imaging with higher resolution for safety and medical applications", identif. code ME 953, Grant agency: ME, 2007-2009, budget:1.4 mil. Kč, role: principal investigator
- “Scintillator performance: quantum effects and delayed recombination”, identif. code M100101212, Grant agency: AVČR, 2012-2015, budget: 0.8 mil. Kč, role: principal investigator
- “Inorganic scintillators: novel synthesis and size-dependent characteristics”, identif. code GA13-09876S, Grant agency: GAČR, 2013-2016, budget Fzú: 4.15 mil. Kč, role: principal investigator
- “Synthesis, characterization and tailoring the properties of luminescence nanocomposites“, identif. code GA17-06479S, Grant agency: GAČR, 2017-2019, budget Fzú: 3.15 mil. Kč, role: principal investigator

Obtained grants/ international:

- “NATO Science for Peace grant” identif. code: 973510-Scintillators, (2000-2003), total budget: cca 8 mil. Kč, role: team member
- EU INTAS, identif. code: 04-78-7803, 2005-2007, total budget 2.1 mil. Kč;
- Partner of EU FP7 project “Luminet”, FP7-PEOPLE-2012-ITN, identif. code: 316906 (2012-2016), budget for FZU: 5.5 mil. Kč, role: team member
- Partner in Horizon 2020 project INTELUM, H2020-MSCA-RISE-2014, identif. code: 368921 (2015-2018), budget for FZU: cca 1 mil. Kč, role: team member
- H2020-TWINN-2015 project ASCIMAT, identif. code: 690599, 2016-2018, budget for FZU: 13 mil. Kč, role: team member

Invited talks at international conferences:

1. E. Mihokova, M. Nikl, S. Baccaro, A. Cecilia: *Scintillator and phosphor materials: latest developments and applications*, Astroparticle, particle and space physics, detectors and medical physics applications, Como, Italy, 2005
2. E. Mihokova, L. S. Schulman, The role of breathers in anomalous decay of luminescence, International symposium on dynamical properties of solids, Český Krumlov, ČR, 2005
3. M. Nikl, A. Yoshikawa, A. Vedda, V. Laguta, E. Mihokova, J.A. Mares, K. Nejezchleb, G. Ren and K. Blazek, *Bottlenecks in scintillation mechanism of complex oxide scintillators*. ISLNOM-5, Pisa, 2009

Teaching and supervision:

Clarkson University, Potsdam, USA- „Quantum mechanics I“, 2003
 Clarkson University, Potsdam, USA- „Solid State Physics I“, 2003
 University of Milano-Bicocca, Italy, Luminescence and scintillating materials, 2009

FJFI ČVUT – „Solid state theory I“ 2014-2017

Supervisor of 2 Diploma thesis and 1 PhD thesis as supervisor specialist

Publication activity (after Web of Science):

h-index = 34

Total number of publications =204

Total number of citations (without selfcitations): 4018

6 Selected publications

1. L. S. Schulman, E. Mihokova, A. Scardicchio, P. Facchi, M. Nikl, K. Polak, and B. Gaveau: Slow relaxation, confinement and solitons. *Phys. Rev. Lett.* **88**, 224101 (1-4) (2002).
2. M.Nikl, E.Mihokova, Z.Malkova, A.Vedda, M.Martini, K.Shimamura and T. Fukuda: Ce^{3+} luminescence in a $LiBaF_3$ single crystal at low temperatures. *Phys. Rev. B* **66**, 184101-(1-5) (2002).
3. L. S. Schulman, D. Tolkunov, E. Mihokova, Stability of quantum breathers, *Phys. Rev. Lett.* **96**, 065501 (1-4) (2006).
4. E. Mihokova, M. Nikl, M. Bacci, M. Dusek, and V. Petricek, Assignment of 4f-5d absorption bands in Ce-doped $RAIO_3$ ($R=La, Gd, Y, Lu$) perovskites, *Phys. Rev. B* **79**, 1951309(1-7) (2009).
5. E. Mihokova, N. Chiodini, M. Fasoli, A. Lauria, F. Moretti, M. Nikl, V. Jary and A. Vedda, Intrinsic and impurity-induced bands in $SrHfO_3$. *Phys. Rev. B* **82**, 165115(1)-165115(7) (2010).
6. E. Mihokova and L. S. Schulman, Discrete breathers and the anomalous decay of luminescence, *J. Phys. A: Math. Theor.* **43**, 183001 (1-46) (2010). Topical review.
7. E. Mihokova, L. S. Schulman, V. Jary, Z. Docekalova, M. Nikl, Quantum tunneling and low temperature delayed recombination in scintillating materials, *Chem. Phys. Lett.* **578**, 66-69 (2013).
8. E. Mihokova, V. Jary, L. S. Schulman, M. Nikl, Delayed recombination and excited state ionization of the Ce^{3+} activator in the $SrHfO_3$ host, *Phys. Stat. Sol. RRL* **7**, 228-231 (2013).
9. L. Havlak, J. Barta, M. Buryi, V. Jary, E. Mihokova, V. Laguta, P. Bohacek, M. Nikl, Eu^{2+} stabilization in YAG structure: Optical and electron paramagnetic resonance study, *J. Phys. Chem. C* **120**, 21751-21761 (2016).
10. R. Kral, V. Babin, E. Mihokova, M. Buryi, V. V. Laguta, K. Nitsch, M. Nikl, Luminescence and charge trapping in Cs_2HfCl_6 single crystals: Optical and magnetic resonance spectroscopy study, *J. Phys. Chem. C* **121**, 12375-12382 (2017).