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Review Article

Deep Level Saturation Spectroscopy

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We review the "Deep Level Saturation Spectroscopy" (DLSS) as the nonlinear method to study the deep local defects in semiconductors. The essence of a method is determined by the processes of sufficiently strong laser modulation (up to saturation) of quasistationar two-step absorption of the probe light via deep levels (DLs). DLSS is based on nonequilibrium processes of the optically induced population changes for deep levels which lead to the changes in an impurity absorption. This method allows us the separation of the spectral contributions from different deep centers (even in the case of their full spectral overlap), on the basis of the difference of their optical activity (photon capture cross-sections) and of their electroactivity difference (carriers capture coefficients). As shown, DLSS is allowed to determine directly the main set of phenomenological parameters (cross-sections, concentration, bound energy, etc.) for deep local defects, their content and energy position in the band gap. Some important aspects of DLSS were shown also: the possibility to connect directly the measured data to the local centers which are participating in radiative recombination, and also the possibility to study directly the phonon relaxation processes in the localized states of deep defects.

1. Introduction

Optoelectrical properties and the applications of semiconductors, accordingly, are significantly defined by the defects, created from stoichiometry deviations, and by the presence of impurities, including uncontrollable ones.

Well-known "Deep Level Transient Spectroscopy" (DLTS) [1] is a sensitive method to study deep levels (DLs) and is widely used for measurements of an electrically active defects in semiconductors. It is based on the temperature scanning of the capacitance transient of a reverse-biased barrier and allows determining the activation energy and concentration of the traps, also thermal emission rates and the capture cross-sections. There are many more varieties, like, for example, "Deep Level Optical Spectroscopy" (DLOS, [2]), or "Acousto-electric DLTS" [3]. The resolution of DLTS can be substantially increased when the so-called "Laplace DLTS" is used [4, 5].

The development of nonlinear optics exposes unique possibilities, partially of two-photon spectroscopy [6–10] as the investigation tool of energetic structure of bands, elementary excitations, and local defects in crystals.

The intrinsic interband two-photon absorption (TPA) is known to be a fundamental process with participation of the crystal native band states as initial and final, as well as intermediate virtual states of a TPA event [6–10]. It is coherent and inertialess and does not saturate.

A lot of defects, resonant with exciting radiation which changes their population, give rise to incoherent two-step absorption via such deep levels (DL) as real intermediate states [11–15]. Laser modulation of TSA consists of saturable and inertial processes, and their spectral influences are specifically variable by different DLs competition.

Below the theoretical basis of other, nonlinear spectroscopic technique, our named as "*Deep Level Saturation Spectroscopy*" (DLSS), based on the nonequilibrium processes of optically induced changes in impurity absorption, and showing unique opportunities for the study of deep levels in semiconductors [11–15], is presented. Related researches were spent also in [16, 17] and, however, were limited to qualitative interpretation of the experimental data.

In [18, 19] we reported a direct investigation of local electron-phonon interaction effects by the DLSS method in ZnS : Cu crystals. It was shown experimentally that DLSS is

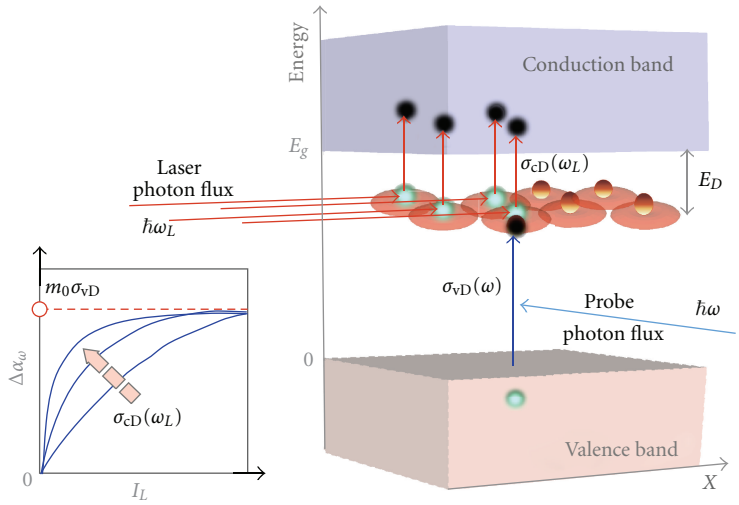

FIGURE 1: Illustration of the "light impact" effect of the donor states depopulation by the laser pulses, completely emptying donors during the irradiation at sufficient density of photons. A probe beam $\hbar\omega$ tests the growth of induced absorption $\Delta\alpha$ due to an emptying of the centers by laser pulses with quanta $\hbar\omega_L$. Intensity dependence $\Delta\alpha(I_L)$ is saturated with I_L growth (an insert).

an exceptionally efficient technique for direct studies of the phonon relaxation processes in the localized states of deep defects.

DLSS method can be divided into the two complementary techniques, differing by the character of the modulation of deep levels population. Firstly we shall discuss in Section 3 the effects of "direct modulation" by photoionisation of DL when defects are characterised on the basis of their optical activity. Then, in Section 5, the consideration of the effects of "indirect modulation" of DL population through the capture of the nonequilibrium carriers generated by interband two-photon absorption is given. As a result, additional parameters of deep levels based on their electro-activity are determined.

The key idea of a method is based on the *"light impact"* effect, as the influence of the short and sufficient powerful laser light pulses, acting as a "photons blow," to the ensemble of DL, completely emptying or filling their states (population saturation) during modulation (Figure 1). The term "light impact" has been entered earlier, studying defects from photoconductivity dynamics [20, 21].

It is normal to assume that during the moments of the laser light "impact" ($I_L(t)$—photon flow density),—only photoionization of DL (with cross-section $\sigma_{cD}(\omega_L)$) by such photons $\hbar\omega_L$ is essential, changing their population $m(t)$ according to the simple rate equation

$$\frac{dm(t)}{dt} = -m(t)\sigma_{cD}(\omega_L)I_L(\omega_L, t), \tag{1}$$

written here for the "donor → conduction band" transitions. Whence we get depopulation changes for the donor states

$$\begin{aligned} m(t) &= m_0 \exp\left(-\sigma_{cD}\int_0^t I_L(t')dt'\right) \\ &= m_0 \exp\left(-\sigma_{cD}I_L^{\Sigma}\right), \end{aligned} \tag{2}$$

and, accordingly, the induced changes in absorption of probing light flow, shown by blue arrows in Figure 1 (see (6))

$$\Delta\alpha(\omega; t, I_L) = m_0\sigma_{vD}(\omega)\left[1 - \exp\left(-\sigma_{cD}^{L}I_L^{\Sigma}\right)\right]. \tag{3}$$

So, at the certain intensities I_L of the modulation, defined by the cross-section of laser photon capture $\sigma_{cD}(\omega_L)$, further intensity growth does not increase the induced absorption of probe light $\hbar\omega$ since the centers are emptied completely by intense modulation. It is illustrated by the fan of saturated dependences on light intensity (3) shown by inset in Figure 1.

Character of the saturation in (3) is defined only by the value of DL photoionization cross-section $\sigma_{cD}(\omega_L)$, that is, by their "optical activity." Namely, the bigger value of this parameter is, the more quickly a full emptying of DL is reached, and consequently the saturation of induced absorption $\Delta\alpha$. This phenomenon enables direct measurement of this initial parameter $\sigma_{cD}(\omega_L)$ for DL. Below we have shown that discussed saturation effects enable us to determine the full set of DL parameters. This specificity also defines the choice of our abbreviation: *"DLSS" DL saturation spectroscopy*.

The essence of a DLSS method consists in the following. During the two-step absorption (TSA) of probe quanta from transparency area ($\hbar\omega < E_g$) of media, there is a consecutive excitation of electrons from a valence band to conduction band through the deep local states in the forbidden gap. This can be classified as *quasistationary TSA* ($v \to D \to c$) for the probe light beam with long enough illumination ($\Delta t > \tau$, τ—lifetime of localized carriers) and *nonequilibrium TSA* ($\Delta t_L < \tau$) for the mixed "probe + laser" two-step transitions $v \to D \to c$. At the combination of long probe and short laser light pulses (Figure 2), so-called, laser modulation of TSA (LM TSA) takes place [11, 14, 22–26], which possesses the necessary spectroscopic opportunities.

Unlike the coherent two-photon absorption (TPA) [23, 26–28] via virtual states, each step of TSA is caused by the combination of one-photon transitions which change

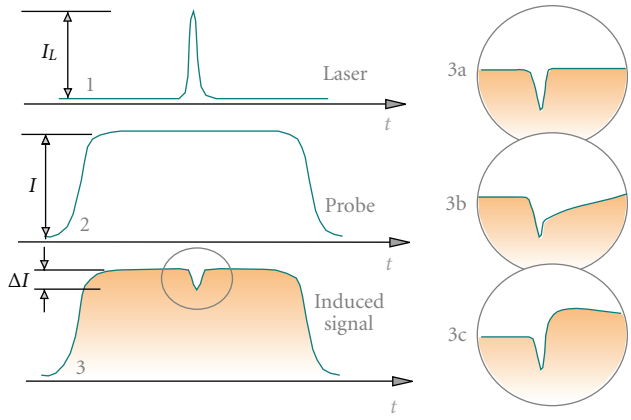

FIGURE 2: Scheme of the typical for DLSS experiment signals: short modulating laser light pulse (1), long probe light pulse (2), and resulting induced absorption signal (3) after crystal transfer. Insets (3a)–(3c) show detailed signals of different nature: coherent inertialess two-photon absorption (3a), combinations of TPA, and laser modulation of incoherent two-step absorption (induced absorption (3b) and bleaching (3c)).

the population of the real intermediate local states. Consequently, the dependence of the induced response on their filling dynamics (population depopulation) is appeared. It concerns to the distinctive attributes of the incoherent processes.

Scheme of the typical for DLSS experiment signals are shown in Figure 2. Insets here show detailed signals of different nature: coherent inertialess two-photon absorption (3a) and combinations of TPA and incoherent LM TSA (induced absorption (3b) and bleaching (3c)). According to Figure 2, the values of induced absorption are determined as

$$\Delta\alpha = \left(\frac{\Delta I}{I}\right)\frac{1}{I_L d}, \tag{4}$$

where d is length of the sample.

In Section 5 it is shown that if laser quanta energy is increased so that interband two-photon excitation appears ($\hbar\omega_L > E_g/2$), then it results additionally in the effects of photoinduced absorption due to the changes in deep level population caused by the capture of nonequilibrium free carriers. These signals (γ-DLSS) are defined by the coefficients γ of the carrier capture and compete with (σ-DLSS) signals discussed before due to σ-photoionization of DL. This specificity expands opportunities to full metrology of opto-, and electroactivity of DL's in semiconductors. As a result, two complementary DLSS methods, differing by the character of DL population modulation, may be realized.

The transient DLSS method was used successfully in studying the properties of DLs in ZnTe [12], ZnSe [11, 25], CdS [29], ZnO [30], and ZnS [18, 19] wide-bandgap crystals. Due to its realization specific, such as necessity to use a thick enough samples (1–10 mm), only deep enough local defects (with binding energies > 100 meV) may be evident and characterised by DLSS.

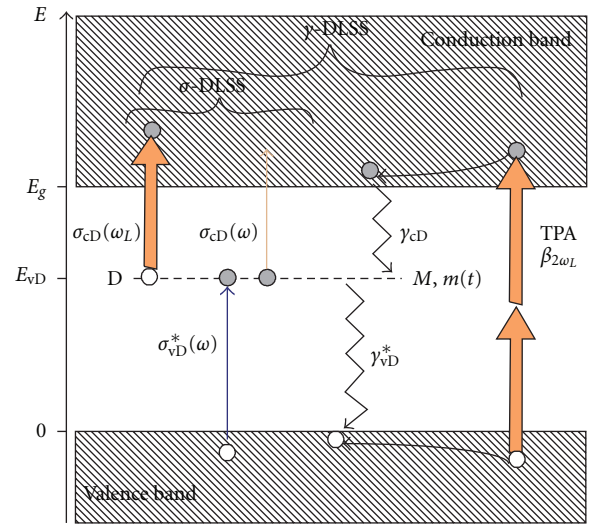

FIGURE 3: Scheme of optical transitions for DLSS calculations in the case of donor (D) states. Fat arrows, DL photoionization $\sigma_{cD}(\omega_L)$ transitions with laser quanta absorption, and interband two-photon (TPA) excitation of a crystal $\beta(2\omega_L)$. Thin arrows, probe light absorption. Broken lines, free carriers capture.

2. DLSS Theory

Let us outcome a theoretical support of the presented method, which is defined by the phenomenon of deep laser modulation, up to saturation, of the quasistationary two-step absorption of the probe light beam via the deep centers in the bandgap.

2.1. Two-Step Processes via Deep Centers and Their Laser Modulation. Let's consider optical transitions (Figure 3) through the deep donor states D with energy E_D, concentration M, and quasistationary electron population $m_0 \equiv m(0)$, established by TSA of probe light $\hbar\omega$ before the action of laser modulation (LM). Last value m_0 defines the initial absorption of probe light

$$\alpha(\omega, 0) = m_0\sigma_{cD}(\omega) + (M - m_0)\sigma_{vD}(\omega), \tag{5}$$

and then, after the action of pulsed modulation $I_L(t)$, we shall get the same equation for $\alpha(\omega, t)$, but now with $m(t)$. Here $\sigma_{vD}(\omega)$ and $\sigma_{cD}(\omega)$ are the capture cross-sections for probe photons $\hbar\omega$ in the transitions "valence band → donor" (*photoneutralization*) and "donor → conduction band" (*photoionization*), accordingly.

It follows, that laser light-induced absorption $\Delta\alpha(\omega)$ of a probe light, counting from the moment of laser switch-on $t = 0$ ($m(t = 0) = m_0$), is defined as

$$\begin{aligned}\Delta\alpha(\omega, t) &\equiv \alpha(\omega, t) - \alpha(\omega, 0)\\ &= (m_0 - m(t))[\sigma_{vD}(\omega) - \sigma_{cD}(\omega)].\end{aligned} \tag{6}$$

The rate equation for the dynamics of DL population $m(t)$, under transitions shown on Figure 3, will be

$$\frac{dm(t)}{dt} = (M - m(t))\left[\sigma_{\mathrm{vD}}(\omega)I(\omega) + \tau_{\Sigma}^{-1}\right] - m(t)[\sigma_{\mathrm{cD}}(\omega)I(\omega) + \sigma_{\mathrm{cD}}(\omega_L)I_L(\omega_L, t)], \quad (7)$$

where lifetime τ_{Σ} of DL nonequilibrium population is entered as

$$\tau_{\Sigma}^{-1}(t) = \left[\gamma_{\mathrm{cD}}n(t) - \frac{m(t)}{M - m(t)}\gamma_{\mathrm{vD}}p(t)\right], \quad (8)$$

which is defined by the capture coefficients of the free electrons γ_{cD}, and the holes γ_{vD} and by their concentrations $n(t)$ and $p(t)$.

Generally, it is necessary to write down the corresponding equations and neutrality conditions for concentration of free carriers $n(t)$ and $p(t)$, as function of illumination influence. Thus, carriers dynamics will be defined by a competition of number of processes (capture, recombination, etc.) which are defined by a set of unknown centers unessential responsible for TSA. The solution of the system of full equations is practically unreal and assumes the multivariate modeling hiding an essence of processes.

All mentioned problems disappear if we consider "differential" specificity of the discussed experimental setup (Figure 2). *Firstly,* quasistationary population is reserved ($n(0)$, $p(0) = \mathrm{const.}$) during the measurements of $\Delta\alpha(\omega, I_L, t)$, because of invariance of influence to the crystal states of a "white" probe illumination, monochromatised only after passing the crystal. *Secondly*, the induced absorption, according to (6), is registered as small differential signal on the strong "background." As a rule, duration of modulation is essentially less than DL population relaxation times $\Delta t_L < \tau_{\Sigma}$, which is supervised in the measurements from signal kinetics.

Thus, the measured response of the centers unable to react to the changes of carriers number in the bands and it is normal to consider $\tau_{\Sigma} = \mathrm{const.} > \Delta t_L$ since if at maximal modulation intensity I_L the condition $\Delta t_L < \tau_{\Sigma}$ is kept, then at any smaller I_L it will be kept also (see (8)). This condition can be checked up experimentally from supervision of the induced absorption signal kinetics. Thus, free carrier's relaxation solution necessity is neglected, but for the population of the centers it is solved easily.

Going from the similar modeling, two DLSS methods σ- and γ-DLSS, differing by character of laser influence on DL population and giving the complementary information, are realized experimentally.

3. Regime of Direct Modulation by Laser Photoionization (σ-LM TSA)

Direct laser modulation of DL population is realised by their photoionization, when $\hbar\omega_L > E_M$ and $\hbar\omega_L < E_g/2$. The second condition defines the absence of two-photon excitation of a crystal ($\Delta n, \Delta p \approx 0$; $\tau_{\Sigma} = \mathrm{const.}$), and also the absence of photoneutralization of the centers by laser quanta ($\sigma^*(\omega_L) = 0$).

The scheme of optical transitions in such conditions (σ-DLSS) is presented on the left part of Figure 3. Generalizing (7) for any type of the centers (donors D or acceptors A), we shall obtain that

$$\frac{dm(t)}{dt} = MG - m(t)[G + R + \sigma(\omega_L)I_L(t)]. \quad (9)$$

Here new designations are entered

$$G \equiv \sigma^*(\omega)I(\omega) + \sigma^*(\widetilde{\omega})I_0(\widetilde{\omega}) + \tau^{-1},$$
$$R \equiv \sigma(\omega)I(\omega) + \sigma(\widetilde{\omega})I_0(\widetilde{\omega}) + \tau^{*-1}, \quad (10)$$

where the index $*$ points out the processes of carriers exchange with the band, more distant energetically from the local levels; $\tau = 1/(\gamma \cdot n)$—localized carrier lifetime relating to the carrier capture with capture coefficient γ. The influence of an additional stationary illumination I_0 controlling initial DL population m_0, according to the quasistationar solution of (9) at $I_L(0) = 0$, is entered as

$$m_0 = \frac{MG}{G + R}. \quad (11)$$

The solution of (9) gives us the dynamics of DL population $m(t)$ that is

$$m(t) = Z^{-1}(t)\left[m_0 + MG\int_0^t Z(t')dt'\right], \quad (12)$$

$$Z(t) \equiv \exp\left[(G + R) + \sigma(\omega_L)\int_0^t I_L(t')dt'\right]. \quad (13)$$

Here is important the fact that during the influence of a laser pulse all other disturbing factors can be unconsidered. Due to a small duration and high intensity of laser pulses, the main mechanism of DL occupation changes is the emptying of the centers by laser quanta. This extremely simplifies calculations. This is used for (1)–(3). So, the progress of induced absorption of probe light during its laser modulation is defined by (6) and by the solution of (7) with dominating component $\sigma(\omega_L)I_L$ [11, 12, 15, 26]

$$\Delta\alpha_{\sigma}(\omega; t, I_L) = m_0[\sigma_{\mathrm{vD}}(\omega) - \sigma_{\mathrm{cD}}(\omega)] \cdot \left\{1 - \exp\left[-\sigma_{\mathrm{cD}}(\omega_L)\int_0^t I_L(t')dt'\right]\right\}. \quad (14)$$

This expression defines the basic regularities of σ-DLSS, illustrated by Figure 4 where formation of induced absorption spectra is shown.

(a) *Spectra* of DLSS are defined by a difference of the spectra of photoneutralization and photoionization cross-sections $\Delta\alpha(\omega) \sim \sigma_{\mathrm{vD}}(\omega) - \sigma_{\mathrm{cD}}(\omega)$. It follows that $\Delta\alpha(\omega)$ spectra give us DL energy location in a forbidden gap relative to both c and v bands. It is defined (Figure 10) from the long-wave thresholds of induced bleaching ($E_{\mathrm{cD}} = E_M$) and induced absorption bands ($E_{vD} = E_g - E_M + \Delta_S$), where Δ_S is the Stokes losses.

Let's discuss the formation of the σ-DLSS spectra (Figure 4). Deep levels at weak illumination are manifested

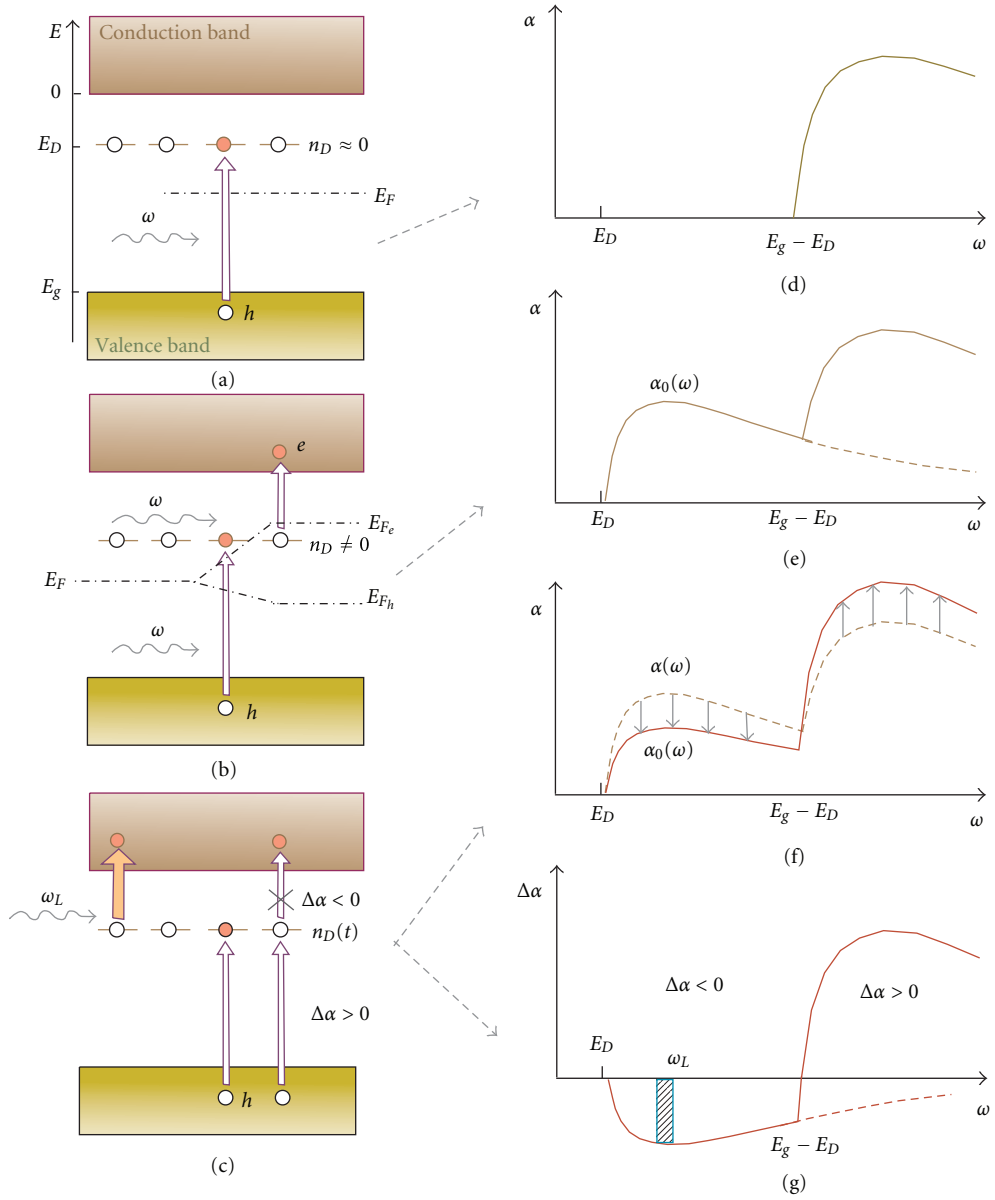

FIGURE 4: Schemes of stationary $\alpha(\omega)$ and induced $\Delta\alpha(\omega)$ impurity absorption spectra formation and of the optical transitions, explaining σ-DLSS: regime of laser modulation of TSA by photoionization, at the absence of TPA-generation ($2\hbar\omega_L < E_g$). (a) and (d). At small intensity, "dark" filling of the compensated donors, only the band of photoneutralization is observed. (b) and (e) At the intensities of probe light, breaking a dark-filling of DL's, photoionization band appears. (f) Changes of TSA spectra as a result of laser modulation. (c) and (g) Induced absorption spectrum.

in absorption spectroscopy by transitions to only one of the bands. This is either the valence or the conduction band [18]. The type of the transitions (ionization or neutralization) is determined by the steady-state occupation of DLs or by the degree of compensation of the crystal, that is, the position of the Fermi level with respect to the energy levels of the defects. Consequently, the same centers can differently be evident in different samples of crystals and never revealing their "full" spectra. For a case Figures 4(a) and 4(d) DL's are compensated and expose only in the photoneutralization process. Thus, it is hardly possible to investigate the full absorption spectrum by traditional methods of steady-state spectroscopy. By the word "full" t the possibility of simultaneously detecting both neutralization "$v \rightarrow D$" and ionization "$D \rightarrow c$" transitions is mean.

During transitions, the optical charge exchange of local centers, that is, lattice relaxation in the vicinity of the defect proceeds simultaneously. In other words, localized carriers generate phonons. This is described by the Franck-Condon rule of Stokes losses. In [18, 19] it was shown that the given specificity of DLSS method is exclusively effective for the direct studies of phonon relaxation effects of local states.

The "full spectrum" of DLs may be measured only by the transient spectroscopy techniques, using additional pulsed illumination, such as DLSS. Then, a nonequilibrium partial occupation of the defect states is created. Figure 4 illustrates the formation of the photoinduced spectrum $\Delta\alpha_\sigma(\omega)$ (14) in the presence of compensated deep donors D. Here, Figures 4(a) and 4(b) demonstrate the appearance of two-step transitions, and Figure 4(c) their laser modulation. The initial part of the photoneutralization spectrum 4(d) and the full TSA spectrum 4(e) are given here. The reaction of two-step transitions 4(b) and their spectrum 4(e) to additional modulation by a laser pulse $\hbar\omega_L$ is shown 4(f) as the changes in the full absorption spectrum $\alpha(\omega)$ and 4(g) as the measured induced absorption spectrum $\Delta\alpha(\omega)$.

As a result of a longtime ($\Delta t \gg \tau$) probe light TSA, the quasistationary filling of DL with electrons m_0 = const. is established (Figure 4(b)). It is defined by a ratio of the transition intensities for the both TSA steps and by reverse capture of the carriers (Figure 4(e)). It corresponds to the introduction of a Fermi quasilevels E_F for holes and electrons. Thus, DL's are exposed in both of the TSA steps, photoionization and photoneutralization (Figure 4(e)).

Because for one of the TSA steps DL is as an initial, and for the other as a final state, the TSA bands react differently to the population changes by the modulation (Figure 4(f)). Photoionization steps ($\sim m_0\sigma(\omega)$) are characterized by the induced transparency ($\Delta\alpha < 0$), and the steps of photoneutralization ($\sim(M - m_0)\sigma^*(\omega)$), by the induced absorption ($\Delta\alpha > 0$). In spectral part $E_M < \hbar\omega < E_g - E_M$, we have $\sigma^*(\omega) = 0$, and only the bleaching of a crystal is observed. At the shorter waves $\hbar\omega > E_g - E_M$ is an actual overlapping and competition of both an induced bleaching (broken line in Figure 4(g)) and the induced darkness signals (see also Figure 10).

(b) *Light-intensity dependences (LID),* or $\Delta\alpha(I_L)$, have satu-ration character ($\Delta\alpha$ at $I_L \to \infty$ does not exceed $\Delta\alpha(\infty) = m_0[\sigma_{vD}(\omega) - \sigma_{Dc}(\omega)]$ according to (14)), that is related with a full emptying (filling) of the donor (acceptor) centers under I_L growth.

With the increase of modulation intensity, signals $\Delta\alpha(I_L)$ pass into saturation by complete laser blenching of DL. The saturation character is determined only by a cross-section value of DL photoionization $\sigma_{cD}(\omega_L)$, that is, by "optical activity" of the centers. The higher $\sigma(\omega_L)$ leads to more effective DL emptying and $\Delta\alpha_\sigma(I_L)$ is saturated at less I_L.

The LID's $\Delta\alpha(I_L)$ in Figure 5(a) were shown for the different photoionization cross-sections $\sigma_i(\omega_L)$. The sequence of the curves 1–3 corresponds to the reduction of $\sigma_i(\omega_L)$ values by the order. It shows high enough accuracy in definition of the $\sigma_i(\omega_L)$ parameters.

The amplitude of $\Delta\alpha_\sigma$ in saturation is determined by quasistationary DL occupation m_0. Curves 1, 2′, and 3′ in Figure 5(a) show us the LID changes with the growth of initial population m_0 of the centers.

Thus, the parameter $\sigma_i(\omega_L)$ defines the character of DL saturation, and the initial population of centers influences only amplitudes of the signals. As a result we have a unique *opportunity to determine separately* $\sigma_i(\omega)$ and m_0 values, which are inseparable in linear optics of defects because it includes them in a product form $\alpha = \sigma m_0$, equal to optical losses.

Therefore we shall change (14) to more convenient form for the experimental data analysis, expressing the laser intensity by its effective duration

$$I_L^a \equiv (\Delta t_L)^{-1} \int_{-\infty}^{\infty} I_L(t)dt. \tag{15}$$

Then the total PIA coefficient β^Σ, reduced to the power density of modulating radiation ($\beta^\Sigma \equiv \Delta\alpha^\Sigma/I_L$), corresponding to the competition of TPA and several channels of σ-DLSS, can be expressed as

$$\begin{aligned}\beta_\sigma^{(\Sigma)}(\omega, I_L^a) &\equiv \frac{\Delta\alpha^\Sigma}{I_L} = \beta^{\mathrm{TPA}}(\omega) + \sum_i \beta_i^\sigma(\omega, I_L^a) \\ &= \beta_{\omega+\omega_L}^{\mathrm{TPA}} + \frac{1}{I_L^a}\sum_i \beta_i^\sigma(\omega, 0) I_{\sigma_i}\left[1 - \exp\left(-\frac{I_L^a}{I_{\sigma_i}}\right)\right],\end{aligned} \tag{16}$$

where $\beta^{\mathrm{TPA}}(\omega)$ is a constant of TPA ($\omega + \omega_L$). The additivity property, taking place at weak effects ($\Delta I \ll I, I_L$), is used here. Also, the *critical intensities* I_σ are entered conditionally. They describe the saturation effects and are defined by $\sigma_i(\omega_L)$ values according to

$$I_{\sigma_i} \equiv \frac{1.6 \cdot 10^{-16}\hbar\omega_L}{\sigma_{cD}(\omega_L)\Delta t_L}. \tag{17}$$

Plots of (16) were shown in Figure 5(b). An initial slope $\beta_i(0)$ of light intensity dependences (broken line in Figure 5(a)) is defined also by DL parameters

$$\begin{aligned}\beta_i^\sigma(\omega, 0) &\equiv \lim_{I_L \to 0} \beta_i^\sigma(\omega, I_L) = \frac{\Delta\alpha_i^\sigma(\omega, 0)}{I_{\sigma_i}} \\ &= m_i(0)[\sigma_i^*(\omega) - \sigma_i(\omega)]I_{\sigma_i}^{-1},\end{aligned} \tag{18}$$

and it turns out by the extrapolation of experimental LID's to zero intensity I_L. Dimensions: β [cm/MW]; I_L, I_σ [MW/cm^2]; σ [cm^2]; m_0 [cm^{-3}]; Δt_L [ns]; $\hbar\omega_L$ [eV].

It is important to note that Figure 5 illustrates a technique for the separation of spectral contributions to absorption from different by the nature DL's on the basis of the difference of their cross-sections σ_i (Figures 5(a) and 5(b)), and from the difference of their carrier capture efficiency (Figures 5(c) and 5(d)).

(c) *Time development* of a signal $\Delta\alpha(t)$ during laser illumination has an integrated character relating to the envelope of the laser pulse. It is related to accumulation dynamics of localized carriers.

However, at the saturation intensities centers are emptying already at the initial stage of signal development $\Delta\alpha(t)$, as illustrated by Figure 6(a) where calculations of σ-DLSS kinetic at different I_L according to (12) for Gaussian laser pulse (19) are presented

$$I_L(t) = I_L^a \cdot L(t) = I_L^a \exp\left[-\left(\frac{t}{b}\right)^2\right]. \tag{19}$$

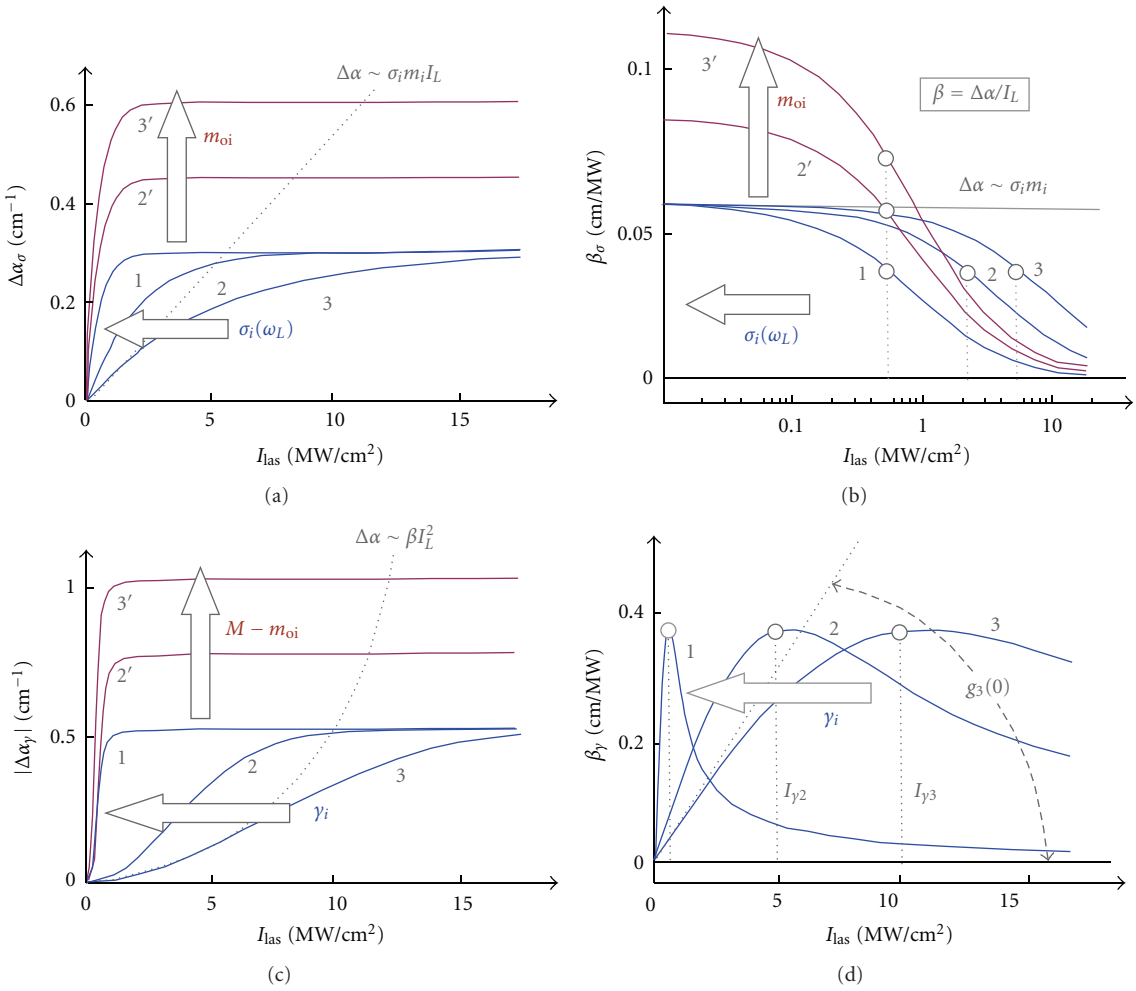

FIGURE 5: Comparison of the light-intensity dynamics of σ-DLSS (laser photoionization of DL, (16)) and of γ-DLSS (28) (capture of nonequilibrium two-photon generated carriers). Separation of the components of different DL absorption bands: (a) and (b) based on the difference of laser photon capture cross-sections σ, that is, on photoactivity of the centers; (c) and (d) based on the difference of coefficients γ of carriers capture, that is, on electroactivity of the centers. The sequence of curves 1, 2, and 3 corresponds to the reduction of capture cross-sections $\sigma(\omega_L)$ for σ-DLSS, or coefficients γ of carrier capture (γ-DLSS). The sequence of curves 1, 2′, and 3′ reflects the influence of initial population m_0 and "vacancies" $M - m_0$ of the centers, accordingly.

Here $b = \Delta t_L/2\sqrt{\ln 2}$, Δt_L is a half-height pulse duration, I_L^a is amplitude, and $L(t)$ is Gaussian envelope of the laser pulse.

(d) *Relaxation* of the induced absorption after the end of modulating pulse, from (6) and (12), will be

$$\Delta\alpha(\omega, t) = \Delta\alpha^a(\omega)\exp[-(G+R)(t-t_k)] \qquad (20)$$

and is defined by the lifetimes τ_Σ (8) of the nonequilibrium population of DL. Here t_k is the moment of the laser pulse termination. It follows that a condition τ_Σ = const., mentioned before, may be checked experimentally from induced absorption kinetics, as independence of relaxation times from intensity of illumination.

4. Excitation Spectroscopy of σ-DLSS: Identification of the Centers as Acceptors or Donors

Let's show an opportunity for the categorization of observable in DLSS centers to the donor or acceptor type. For this purpose the measurements of the excitation spectra of DLSS [15] were based on the fact that the induced bleaching signals were dependent on the spectral composition of the probe beam acting before the laser modulation.

The explanation is given on Figure 7. If the short-wave part of multichromatic probe beam $\hbar\omega_{exc}$, causing photoneutralization σ^*_{cA} of the compensated acceptors in n-type crystals (Figure 7), is blocked, then these acceptors will remain populated completely by electrons. Then, DLSS signal for the quanta $\hbar\omega_{probe}$ is absent since a laser pulse $\hbar\omega_L$ cannot change their population. At the presence of such quanta

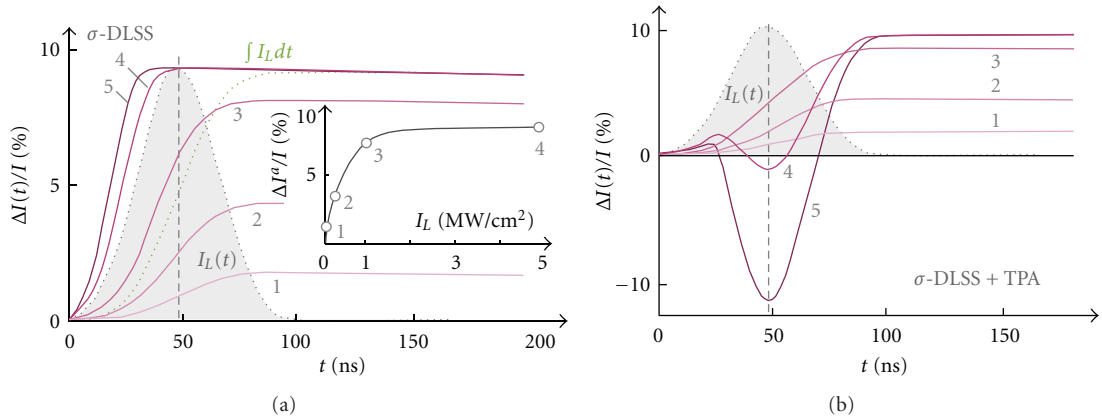

Figure 6: Calculated kinetics of σ-DLSS signals in the spectral area of impurity bleaching ($\hbar\omega > E_g - E_M$) for different levels of modulation I_L; for the cases: without (a) and with account (b) of the coherent TPA contribution. (a) $\Delta\alpha_\sigma(t)$ (12) for a Gaussian laser pulse (19) with $\Delta t_L = 40$ ns (dotted line, integrated envelope $\int_0^t I_L(t')dt'$). Inset-corresponding LID with the numbered points for which the shown same numbered kinetics were calculated ($5 - I_L = 10$ MW/cm^2). Parameters: $M = 10^{17}$ cm^{-3}, $\sigma(\omega) = \sigma^*(\omega) = \sigma^*(\omega_L) = 10^{-17}$ cm^2, $\tau^* = 10^{-5}$ s, $I(\omega) = 10^{21}$ s^{-1}. (b) Similar calculations, but with adding the contribution of an opposite TPA signal ($\beta = 0{,}01$ cm/MW).

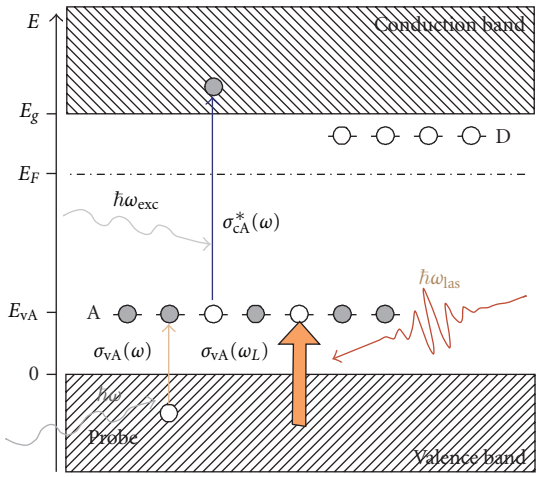

Figure 7: Scheme of optical transitions for the explanation of excitation (stimulation) spectroscopy of σ-DLSS. Acceptors (A) are compensated by shallow donors (D). Fat arrows, transitions of laser photoionization $\sigma_{vA}(\omega_L)$. Thin arrows, absorption of the probe and excitation light.

$\hbar\omega_{exc} > E_{cA}$ in a probe beam (transitions $A \to c$), the centers are partially emptying and bleaching σ-DLSS signals appear.

Thus for the compensated acceptors, the response strongly depends on the spectral composition of preceding illumination and, for the donors, does not depend. The given effects are defined by (11) for the initial centers filling m_0.

We shall consider the maximal value of PIA amplitude $\alpha_{max}(\omega) = \Delta\alpha(\omega, \sigma, I_L \to \infty)$, using (11) and (14)

$$\Delta\alpha_{max}(\omega) = -m(0)[\sigma(\omega) - \sigma^*(\omega)] = \frac{M[\sigma^*(\omega) - \sigma(\omega)](\sigma^*(\tilde{\omega})I_0(\tilde{\omega}) + 1/\tau)}{[\sigma(\tilde{\omega}) - \sigma^*(\tilde{\omega})]I_0(\tilde{\omega}) + 1/\tau + 1/\tau^*}, \quad (21)$$

which defines PIA dependence on the conditions of quasistationary illumination I_0. Assuming small probe intensity I, (21) for the bleaching spectra part becomes

$$\Delta\alpha_{max}(\omega) = -M\sigma(\omega)\frac{\sigma^*(\tilde{\omega})I_0(\tilde{\omega}) + 1/\tau}{1/\tau + 1/\tau^*}. \quad (22)$$

Let's consider two extreme cases:

(a) a weak influence of capture from the neighbor band, that is, $\sigma^*(\omega)I_0(\omega) \gg \tau^{-1}$. It is realized for DL with repulsive potential ($\gamma \ll \gamma^*$) or at their strong compensation ($n_0 \gg p_0$, for acceptors). Thus from (22) we have

$$\Delta\alpha_{max}(\omega) = -M\sigma(\omega)\sigma^*(\tilde{\omega})I_0(\tilde{\omega})\tau^*, \quad (23)$$

whence follows that, at the absence of illumination with $\hbar\omega_{txc} > E_g - E_M$, neutralizing the centers, bleaching signals in the region $E_M < \hbar\omega < E_g - E_M$ are absent in general, since these centers are vacant. Thus, excitation spectrum of DLSS $\Delta\alpha(\omega) = f(\tilde{\omega})$, detecting by the change of illumination quanta energy $\hbar\omega$ at the fixed probe frequency $\hbar\omega_{probe}$, looks like the spectrum of photoneutralization cross-section $\sigma^*(\omega)$ (curve 1 on Figure 8) in the bleaching region $E_M < \hbar\omega_{probe} < E_g - E_M$.

(b) Otherwise, at the domination of the capture from nearest band (uncompensated centers), PIA is independent of the presence of illumination photoneutralizing the centers, or on a spectral composition of probe light (curve 3 on Figure 8).

Corresponding excitation spectra and intensity dependences $\Delta\alpha(I_\omega)$ of DLSS are shown schematically in Figure 8. Thus, from the analysis of DLSS excitation spectra, it is possible to do conclusions about the correlation of capture times τ_{cM} and τ_{vM}, and more, at the known type of dominating conductivity, about the nature of DL's (acceptor or donor?).

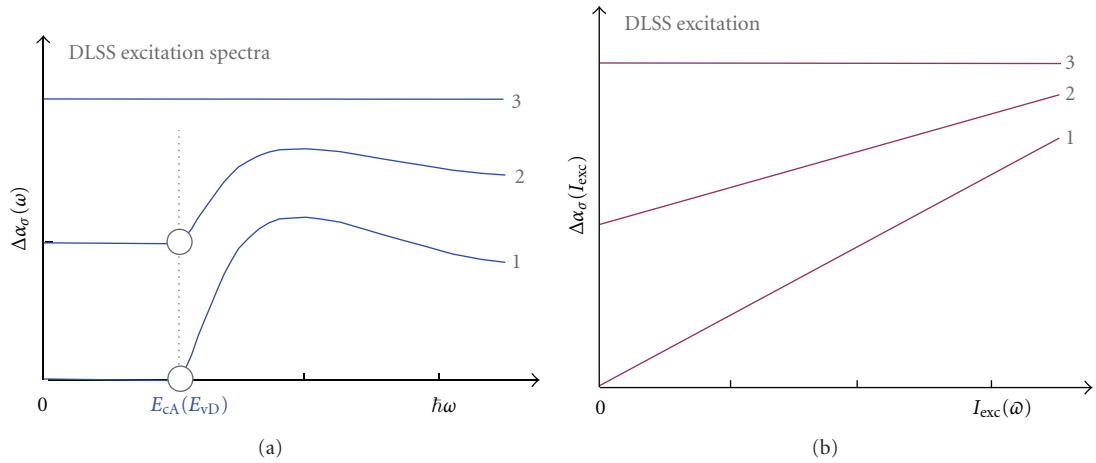

FIGURE 8: Excitation spectra of DLSS (a) and intensity dependences on $I(\omega)$ of the photoneutralizing part of illumination with $\hbar\omega > E_g - E_M$. Ratios of the localized carrier lifetimes: $1-\tau \gg \tau^*$ ($\sigma_{cD} \gg \sigma_{vD}$ or $\sigma_{vA} \gg \sigma_{cA}$, compensated centers); $2-\tau \approx \tau^*$; $3-\tau \ll \tau^*$ (uncompensated centers).

5. Indirect Modulation of TSA from the Capture of Two-Photon Generated Carriers (γ-LM TSA)

Two-photon excitation is volumetric and nonlinear and leads to the effects of absorption by generated nonequilibrium free carriers [12, 28]. The presence in a crystal of the fast carrier capture centers, due to change of their population under TPA excitation, becomes evident in the photoinduced absorption (PIA). This is shown schematically on Figures 9 and 3 (right part).

Such γ-DLSS process is described also by the rate equation (7), where carrier concentration $n(t) = n_0 + \Delta n(t)$ is defined, in difference to (13), before now, by two-photon excitation if $2\hbar\omega_L > E_g$. At short enough laser pulses $\Delta t_L < \tau_r$

$$\Delta n(t) = \Delta p(t) = n(t) - n_0 = \frac{\beta_{2\omega_L} I_L^2}{2\hbar\omega_L} \int_0^t L^2(t')dt', \quad (24)$$

where $\beta_{2\omega_L}$ is TPA constant for laser quanta $\hbar\omega_L$. The concentration of carriers achievable at such excitation does not exceed $10^{17} \div 10^{18}$ cm^{-3}, up to optical breakdown. Neglecting the influence of probe light, balance equation (7) becomes

$$\frac{dm(t)}{dt} = [M\gamma - m(t)(\gamma + \gamma^*)]n(t), \quad (25)$$

and its solution according to (6) gives us PIA coefficient

$$\Delta\alpha_\gamma = [\sigma(\omega) - \sigma^*(\omega)]\left(\frac{\gamma}{\gamma+\gamma^*}M - m(0)\right) \cdot \left\{1 - \exp\left[-(\gamma+\gamma^*)\int_0^t n(t')dt'\right]\right\}. \quad (26)$$

It follows that the form of γ-DLSS spectrum is defined by the ratio of the coefficients of carrier capture from nearest (γ) and distant (γ^*) bands (see Figure 3). As a rule, DL are characterized by primary capture from a near band, that is, $\gamma \gg \gamma^*$ or $\gamma_{cD} \gg \gamma_{vD}$ (Figure 3).

Thus, the substitution of (24) into (26) gives

$$\Delta\alpha_\gamma(\omega; t, I_L) = (M - m_0)[\sigma(\omega) - \sigma^*(\omega)] \cdot \left(1 - \exp\left[-\frac{\gamma^*\beta}{2\hbar\omega_L}\int_0^t\int_0^{t'} I_L^2(t')dt'dt\right]\right), \quad (27)$$

which reflects the basic properties of γ-DLSS.

(a) *Spectral appearance* of γ-DLSS is formed according to the scheme Figure 9. Part of reasoning's in occasion of Figures 9(a) and 9(b) remains the same as for σ-DLSS. Main difference is that modulation of DL population occurs indirectly, through the capture of the carriers injected in a crystal volume optically, by TPA-excitation. The difference of $\Delta\alpha_\gamma(\omega)$ spectra (Figure 10) consists in a fact that the capture of carriers from a nearby band leads to the increase in DL population; therefore, γ-DLSS spectrum Figure 9(b) is expected as inverted in comparison with σ-DLSS spectra (Figure 9(a)).

This is illustrated by experimental data for ZnSe [24] in Figure 11 where mentioned DLSS features are presented for laser modulation with $\hbar\omega_L < E_g/2$ (spectrum 1) and $\hbar\omega_L > E_g/2$ (spectrum 2). Curve 3 shows the calculated spectrum. The intensity dependences $\Delta\alpha(I_L)$ are compared with the theoretical ones according to (16) and (28) in Figures 11(d) and 11(e), correspondingly. Curves 2 and 3 differ in value σ_{vA} by the order and show the precision of its determination. At $\hbar\omega_L > E_g/2$, TPA excitation takes place, and spectra (2) invert in sign without changing the shape.

However, at the greater carrier capture efficiency from the more removed bands (e.g., domination of holes capture by the donors), DL's are emptying, and γ- as σ-DLSS spectra may also have an identical appearance.

(b) *Light-intensity dependences* of σ- and γ-DLSS are essentially different (Figure 5), that also enables the separation of different centers contributions and their parameters definition.

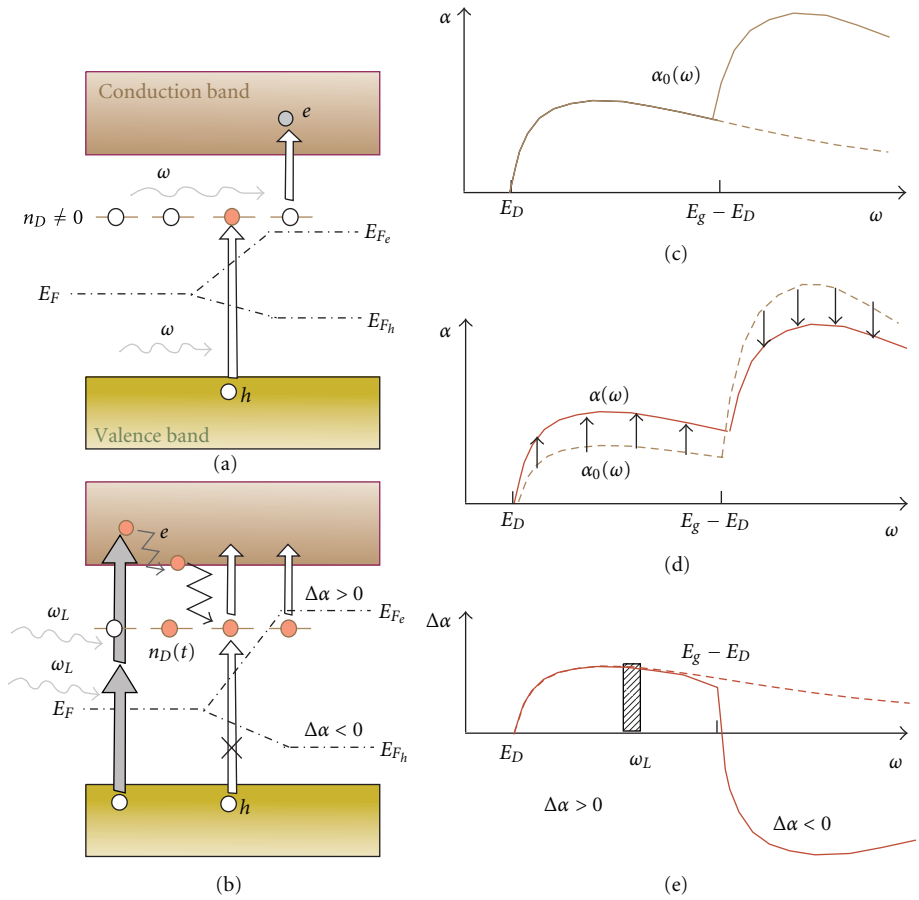

FIGURE 9: Schemes of the spectra formation for stationary $\alpha(\omega)$ and induced $\Delta\alpha(\omega)$ impurity absorption and the optical transitions, explaining γ-DLSS, as the consequence of laser modulation of the quasistationary TSA by the capture of two-photon generated carriers. (a) and (c). Energy scheme and TSA spectrum $\alpha(\omega)$ before the modulation [same as on (b) and (e) parts of Figure 4]. (b) and (d). Energy scheme and TSA spectra changes as a result of laser modulation. (e) Resultant induced absorption spectrum.

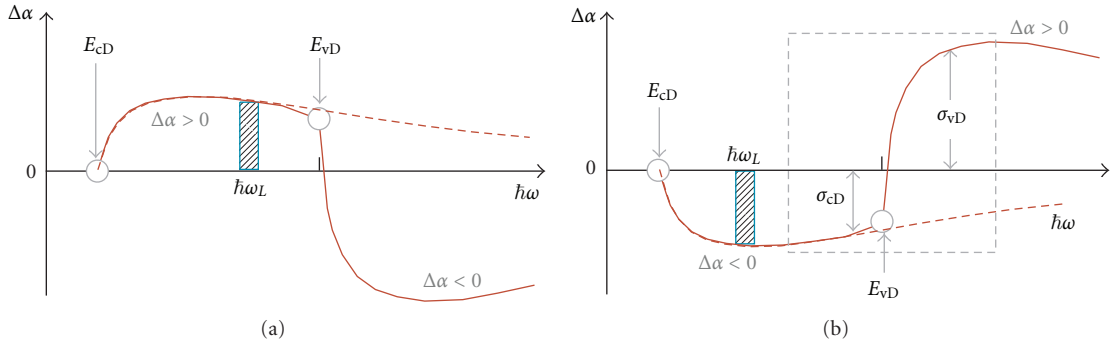

FIGURE 10: Schemes of photoinduced absorption (PIA) spectra $\Delta\alpha(\omega)$ for σ-DLSS (a) and γ-DLSS (b). $\Delta\alpha(\omega)$ give us energy position of DL concerning to both bands, defined from long-wave thresholds of an absorption (E_{vD}) and the bleaching (E_{cD}) bands.

At the small intensities of modulating radiation, PIA have square-law LID (dotted line in Figure 5(c)), following the concentration (24) of two-photon generated carriers. With the growth of excitation, when the number of the vacant centers became low, LID aspires to the saturation (Figure 5(c)), and more quickly at the greater γ values.

It is important that γ-LM TSA gives the information on processes of carrier capture by the centers, that is, about their *electroactivity*. Besides amplitude $\Delta\alpha_\gamma(\infty)$, it is defined by the value $M - m_0$. We shall remind that for σ-modulation the amplitude $\Delta\alpha_\sigma(\infty)$ is defined by value m_0. Therefore carrying out the experiments in both modes of modulation allows us

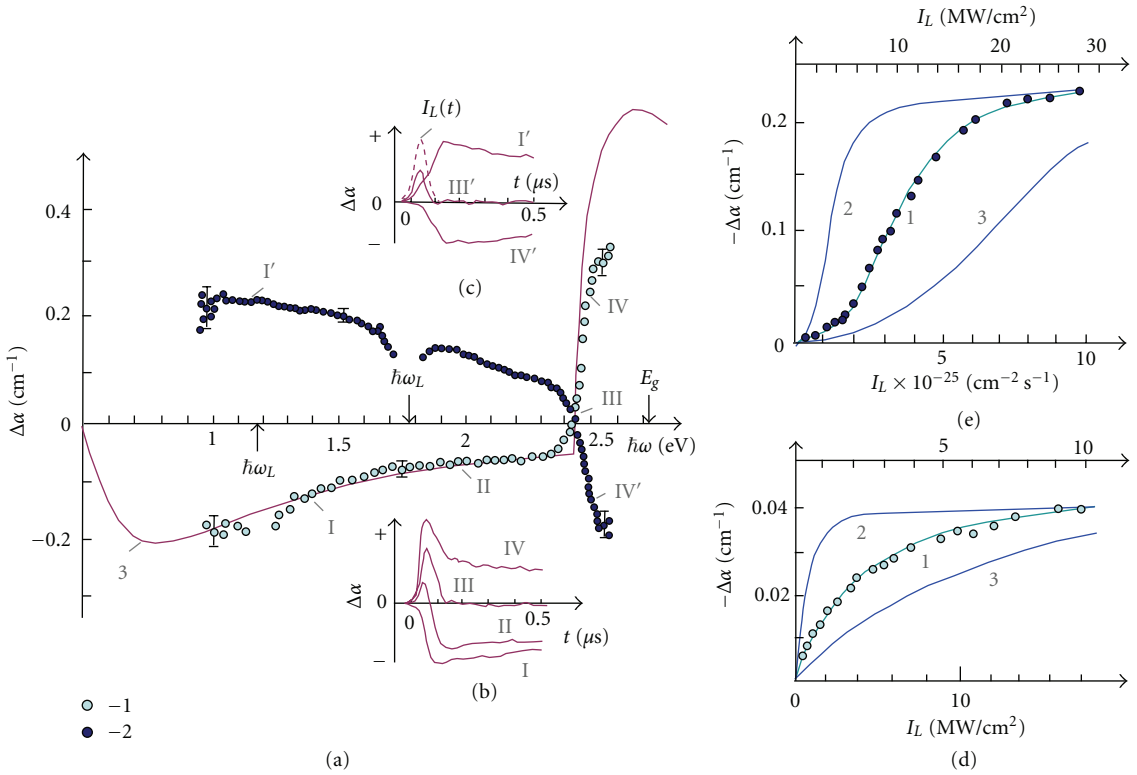

Figure 11: Spectra (a), signals (b) and (c) and dependences on modulation intensity (d) and (e) of the induced absorption of σ-DLSS and γ-DLSS type in ZnSe. The conditions of laser modulation are $\hbar\omega_L = 1.17$ eV $< 1/2E_g$ (a1), (b), (d) and $\hbar\omega_L = 1.78$ eV $> 1/2E_g$ (a2), (c), (e); $\Delta t_L = 30$ ns; $E_g = 2.72$ eV, from [24].

to define directly the concentration of the centers M (32), and also their quasistationary population m_0/M (31).

Let's transform (27) in the same way as(16)

$$\beta^{(\gamma)}(\omega, I_L) \equiv \frac{\Delta\alpha^{\gamma}}{I_L} = \beta^{\mathrm{TPA}}(\omega) + \sum_i g_i(\omega, I_L)$$
$$= \beta^{\mathrm{TPA}}_{\omega+\omega_L}(\omega) + I_L^{-1} \sum_i g_i(\omega, 0) I^2_{\gamma_i} \left[1 - \exp\left(-\frac{I_L^2}{I^2_{\gamma_i}}\right)\right], \tag{28}$$

where experimentally defined values depending on the DL parameters are entered

$$g_i(\omega, 0) \equiv \lim_{I_L \to 0} g_i(\omega, I_L) = \frac{\Delta\alpha_i^{\gamma}(\omega, 0)}{I^2_{\gamma_i}}$$
$$= (M_i - m_i(0))\left[\sigma_i(\omega) - \sigma_i^*(\omega)\right] I_{\gamma_i}^{-2},$$
$$\Delta\alpha_i^{\gamma}(\omega, 0) = (M_i - m_i(0))\left[\sigma_i(\omega) - \sigma_i^*(\omega)\right],$$
$$I^2_{\gamma_i} \equiv \frac{3.2 \cdot 10^{-7} \hbar\omega_L}{\gamma_i \beta_{2\omega_L} \Delta t_L^2}. \tag{29}$$

Parameters $g_i(\omega, 0)$ and I_γ are as an initial slope of LID (at $I_L \to 0$) and a certain critical intensity of modulation at which saturation effects start (see Figure 5(d)). Dependences (28) are shown in Figure 5(d). Figure 5 gives us the comparison of LID (16) and (28) for σ- and γ-DLSS modes in their variability from DL parameters.

It is important to note that Figure 5 illustrates the technique for the separation of spectral contributions from the different DL's to the induced absorption spectra, based on the difference of photon capture cross-sections σ_i, that is, on the photoactivity of the centers.

(c) *Concentration and population of DL.* We shall note that in both techniques the "binding to the center" is made by the characteristic energies of DL (see Figure 10). The conditions of quasistationary excitation of a crystal (initial population of DL m_0 before the modulation) for both techniques are identical. So, the values of PIA coefficients $\Delta\alpha$ at low modulation, $I_L \to 0$, are determined by an initial population m_0 (see Figure 5(a)) and by the donor vacancy $M - m_0$ (see Figure 5(d)), accordingly

$$\Delta\alpha_i^{\sigma}(\omega, 0) = m_i(0)\left[\sigma^*_{\mathrm{vD}}(\omega) - \sigma_{\mathrm{Dc}}(\omega)\right],$$
$$\Delta\alpha_i^{\gamma}(\omega, 0) = (M_i - m_i(0))\left[\sigma_{\mathrm{Dc}}(\omega) - \sigma^*_{\mathrm{vD}}(\omega)\right]. \tag{30}$$

These dependences also define the form of the spectra in Figure 10. From here it is easy to determine an initial degree of DL population

$$\frac{m_i(0)}{M_i} = \left(1 - \frac{\Delta\alpha_i^{\gamma}(0)}{\Delta\alpha_i^{\sigma}(0)}\right)^{-1}, \tag{31}$$

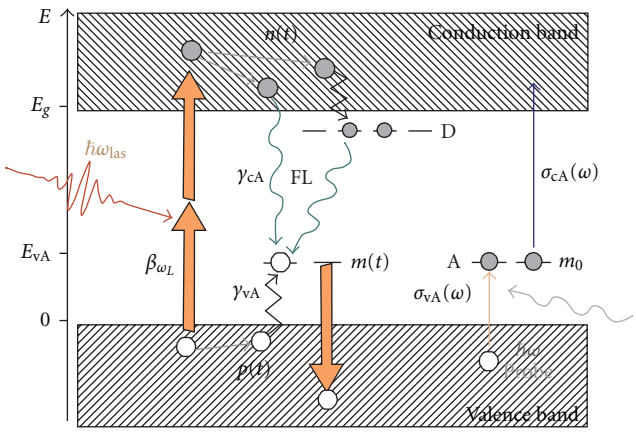

Figure 12: Scheme of the processes considered for the calculations of γ-DLSS dynamics and impurity luminescence.

established during quasistationary TSA of the probe light, before the action of modulation laser pulse, and also to determine the concentration of the centers M

$$M_i = \frac{\Delta\alpha^{\infty}_{\gamma_i}(\omega) - \Delta\alpha^{\infty}_{\sigma_i}(\omega)}{\sigma_{vD}(\omega) - \sigma^{*}_{cD}(\omega)}, \tag{32}$$

where $\Delta\alpha^{\infty}$ is a limiting value of the induced absorption of i-component of DLSS at $I_L \to \infty$, defined from the LID (Figure 5).

6. Correlation between DLSS and Luminescence Dynamics: Identification of Radiation Centers

Metrological opportunities of DLSS in determining the basic set of phenomenological DL parameters are shown above. The problem of identification of the defects nature [13–25] is solved, as usual, from the general principles of technological changes of their content.

Let's show one more aspect of DLSS, an opportunity to connect directly the experimental data with participation of the centers in radiative recombination and to define the luminescence band caused by concrete DL's. Till now, such opportunities have been realized only in the methods of optically detected magnetic resonance (ODMR) [31–34]. Except the information on microscopic properties of the local centers and their physical-chemical identification, ODMR enable also to bind defects to their photoluminescence (PL) bands.

In DLSS for these purposes the effect of dual influence of a laser pulse on the centers population in conditions of TPA excitation and also the dynamics of PL connected with it can be used. It is obvious in "excitation-quenching" effect in PL, illustrated schematically by Figures 12 and 14(b).

The essence of the phenomenon consists with the fact, that the same laser pulse generating free carriers, which are capturing by the centers (γ-DLSS), besides also emptying them simultaneously via photoionization (σ-DLSS). If for this center the second step of recombination is radiative then laser pulse simultaneously also enhances and damps appropriated PL band. The competition of these phenomena develops in time in a complicated manner, but if the carrier lifetimes can exceed the duration of an excitation then PL, it will flare up after laser pulse termination.

Light-intensity dependences (LID) of PL and DLSS are equally defined by the dynamics of the local center population changes Δm, which at the moderate excitation intensities are square laws from I_L, since follows the carrier's concentration. Then with growth of excitation, due to finiteness of the number of local centers, Δm reaches value of their concentration, and PL and DLSS signals are saturated in same manner.

Let's consider dynamics of the discussed processes (Figure 12) from the balance equation (7) for acceptor population changes $m(t)$ at $I_L \gg I$, $\gamma_{vA} \gg \gamma_{cA}$, and $\sigma_{Ac}(\omega_L) = 0$. The solution of (7) then can be expressed as

$$\frac{m(t)}{m_0} = \frac{1}{Z(t)}\left\{1 + \frac{M}{m_0}\gamma_{vA}\int_0^t Z(t')p(t')dt'\right\}, \tag{33}$$

$$Z(t) \equiv \exp\left\{\int_0^t [\gamma_{vA}p(t') + \sigma_{vA}(\omega_L)I_L(t')]dt'\right\}, \tag{34}$$

where m_0 is defined by the quasistationar occupation of an acceptor states due to the probe illumination. The concentration of two-photon generated holes $p(t)$ is defined from (24).

The dynamics of impurity PL for n-type monopolar crystals is described by

$$\Delta I_{lum}(t) = I_{lum}(0) - I_{lum}(t) = \gamma_{cA}n(t)(m_0 - m(t)). \tag{35}$$

The induced absorption at the competition of TPA with σ- and γ-DLSS is described by

$$\Delta\alpha(t) = [\sigma_{cA} - \sigma_{vA}](m_0 - m(t)) + \beta I_L(t), \tag{36}$$

where $\beta \equiv \beta_{\omega+\omega_L}$ is TPA coefficient. The changes of the centers population $m(t)$ (33) with the growth of an excitation I_L are presented in Figure 13(b).

As we see, during the action of laser illumination impurity, luminescence can be blanked completely, sharply flaring up upon the termination of a laser pulse, and then being saturated with growth of excitation. Such analysis was used to explain the behaviour of a G-band of photoluminescence in CdS [29] as annihilation of the excitons, trapped by DA-centers. The effects of the same nature were shown in the laser-induced self-diffraction [35] experiments.

7. Conclusion

The "deep level saturation spectroscopy" (DLSS), based on incoherent nonequilibrium processes of impurity absorption, optically modulated up to saturation [11–15, 22–26], allows to separate the spectral contributions from deep centers of different nature, even in the case when they are

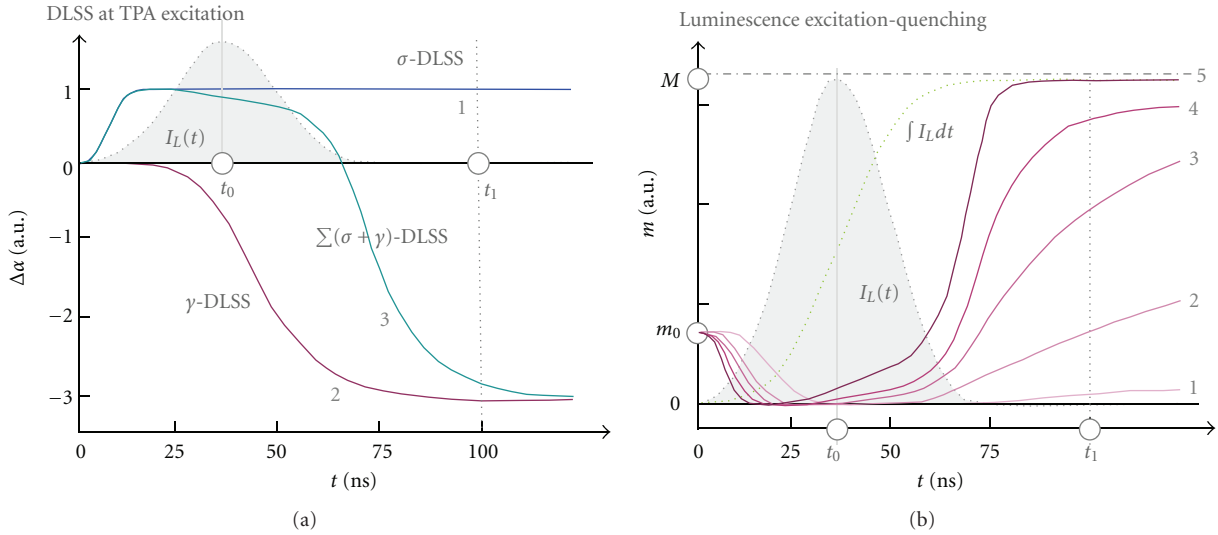

FIGURE 13: Calculation results for a kinetic of DLSS signals (a) and of the centers population (b) by (33) (or PL by (35)) at the two-photon excitation. (a) 1 and 2: $\Delta\alpha(t)$ for the independently proceeding processes of σ- and γ-DLSS, accordingly; 3: induced absorption kinetics at joint action of σ- and γ-DLSS. (b) A kinetic of centers population $m(t)$ at the different intensities of modulation: I_L = 1; 3; 6; 10; 18 MW/cm^2 for curves 1–5, accordingly. M: concentration, m_0: initial population of DL. Dotted line: envelope of a modulation pulse (19). Parameters of CdS are used [24, 29]: $M/m_0 = 4$; $\gamma_{cA} = 10^{-9}$ cm^3/s; $\sigma_{vA}(\omega_L) = 10^{-16}$ cm^2; $\beta = 0,02$ cm/MW; $\Delta t_L = 30$ ns.

spectrally overlapped and irresolvable, and to define directly their basic phenomenological parameters, composition, and the role in the formation of photoelectric properties of crystals [29, 35].

The opportunities of DLSS may be expanded uniquely if two types of laser modulation of TSA, illustrated schematically by Figures 14(a) and 14(b), were realized:

(I) direct modulation of DL population by their photoionization (σ-DLSS) by laser pulse and

(II) "indirect" modulation of DL population by the capture of nonequilibrium carriers (γ-DLSS), two-photon generated by laser pulse. At the conditions of two-photon crystal excitation, both competing σ- and γ-DLSS processes may be actual (Figure 14(c)). Also the appearance of the effects of "enhancement-damping" of impurity luminescence by laser radiation (Figure 14(c)) is possible for defects involved in radiative recombination.

Metrology opportunities of DLSS give a full set of phenomenological parameters of deep centers and their content:

(i) energy position E_{vM} and E_{cM} in forbidden gap,

(ii) values and spectra both of photoionization and of photoneutralization cross-sections: σ_{vM} and σ_{cM},

(iii) coefficients γ_M of dominating carriers capture,

(iv) quasistationary population of the centers, m_0/M,

(v) lifetimes of the localized carriers or times of restoration for quasistationary population,

(vi) concentration of deep centers, M.

Additional features of DLSS method are the following opportunities.

(i) "*Voluminosity" of phenomena.* In DLSS the light beams from the crystal transparency region are "working." It allows to study the defects in the volume of a crystal, including an opportunity of the spatially selective 3D-scannings. The problem of the volumetric homogeneity diagnostics of crystals by content and parameters of local defects may be solved.

(ii) *Opportunity to measure "full spectra" of DL*, that is to register simultaneously the interconnected spectra both as of the photoionization, and of the photoneutralization. This property has been used in [18, 19] for the direct study of the local electron-phonon interaction in ZnS crystals.

(iii) *Separation* of the spectral contributions of the optically competing centers on the basis of their optical activity difference (cross-sections of photon capture; Figure 14(a); σ-DLSS).

(iv) *Separation* of the spectral contributions of the centers on the basis of their electroactivity difference (by carrier capture coefficients; Figure 14(c); γ-DLSS).

(v) *Separation* of the spectral components of defects by the change of the modulation quanta energy, consecutive change of composition of the levels participating in the formation of induced response.

(vi) *Identification* of the centers nature, as donor or acceptor (localization in forbidden gap), from the measurements of σ-DLSS excitation spectra [22].

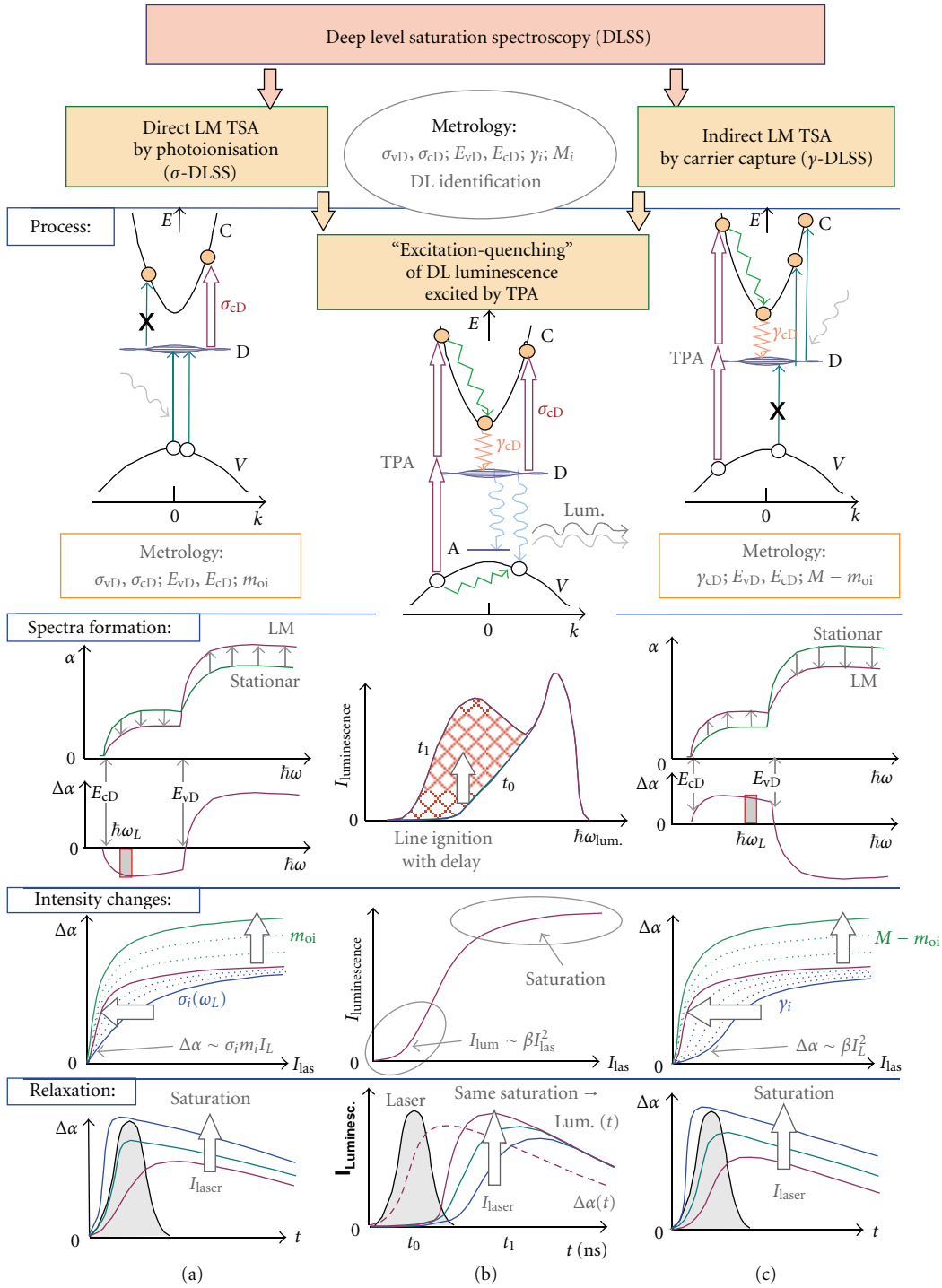

FIGURE 14: Scheme of optical processes of laser modulation of two-step absorption (LM TSA) via deep centers, explaining a method of "deep level saturation spectroscopy" (DLSS). (a) Direct laser modulation of two-step absorption (σ-DLSS) from changes of center population by laser photoionization (σ_M). (b) Competition of σ- and γ-DLSS. Effect of laser "excitation-quenching" of impurity luminescence. (c) Indirect laser modulation of TSA (γ-DLSS) by capture (γ_M) of two-photon generated carriers.

(vii) *Special selectivity to the type of primary carrier capture by the centers*, from the spectral form of γ-DLSS components. Induced response to the carrier capture from a near or distant band, relative to DL level, are different by sign, so the measured spectra are inverted [22].

(ix) *Correlation between the luminescence and induced absorption*. Study of photoluminescence (PL) and its dynamics (effect of laser "excitation-quenching" of PL; Figure 14(b)) and their correlation with induced absorption in the same conditions of two-photon excitation, allows to specify defect

participation in the formation of PL bands [14, 29]. As a result there is an opportunity not only of the defect metrology, but also revealing their participation in the radiative recombination.

The discussed technique is used earlier for CdS [10, 36, 37], ZnSe [11, 13], and ZnO [15, 30] crystals for the study of deep level composition changes due to the crystal technology.

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