Can Transient Phenomena Help Improving Time Resolution in Scintillators?

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***Abstract*—The time resolution of a scintillator-based detector is directly driven by the density of photoelectrons generated in the photodetector at the detection threshold. At the scintillator level it is related to the intrinsic light yield, the pulse shape (rise time and decay time) and the light transport from the gamma-ray conversion point to the photodetector. When aiming at 10 ps time resolution, fluctuations in the thermalization and relaxation time of hot electrons and holes generated by the interaction of ionization radiation with the crystal become important. These processes last for up to a few tens of ps and are followed by a complex trapping-detrapping process, Poole-Frenkel effect, Auger ionization of traps and electron-hole recombination, which can last for a few ns with very large fluctuations. This paper will review the different processes at work and evaluate if some of the transient phenomena taking place during the fast thermalization phase can be exploited to extract a time tag with a precision in the few ps range. A very interesting part of the sequence is when the hot electrons and holes pass below the limit of the ionization threshold. The only way to relax their energy is then through collisions with the lattice resulting in the production of optical and acoustic phonons with relatively high energy (up to several tens of meV) near the ionization threshold. As the rate of such electron/phonon exchange is about 100 events/ps/electron or hole and as the number of electrons/holes generated after mutiplication in a high light yield scintillator like LSO can be as high as 100,000 or more, we end up with an energy deposition rate of about 100 KeV/ps. This energy deposition rate contributes to many fast processes with a characteristic time in the ps range such as band-to-band luminescence, hot intraband luminescence, acoustic shock wave generation, fast local variation of index of refraction, etc. We will discuss if the part of the total energy which is released this way, and which can represent between 50% and 90% of the energy of the incoming ionization radiation, can be efficiently exploited to improve the time resolution of scintillators, presently limited to the 100 ps range.**

***Index Terms*—PET, picosecond, scintillation, scintillator, time-of-flight, transient.**

I. Introduction

THE search and development of scintillators in the last decades has been mainly oriented towards higher light yield and better proportionality in order to improve the energy resolution and shorter decay time to cope with higher event rates while minimizing pile-up.

Recent years have seen the emergence of fast timing capability as a new requirement, mainly driven by High Energy Physics (HEP) and time-of-flight positron emission tomography (TOF-PET) applications.

The quest for rare events in HEP requires the construction of colliders with high luminosity, which is achieved by densely spaced particle bunches separated by only 500 ps in the case of the Compact Linear Collider CLIC [1]. The correct identification of bunches at the origin of an event implies a timing resolution of at least 500 ps for all the recorded by-products of this event (particle tracks or energy deposited in calorimeters). Moreover bunches themselves carry a high density of particles so that each bunch crossing generates several collisions. At the Large Hadron Collider (LHC) at CERN up to 40 pile-up events can be produced at each bunch crossing at the designed luminosity of $2.10^{34}\ \mathrm{cm}^{-2}\mathrm{s}^{-1}$, which will reach 200 pile-up events when the luminosity will be increased to $10^{35}\ \mathrm{cm}^{-2}\mathrm{s}^{-1}$ at the High Luminosity LHC [2]. For a luminosity region of about 10 cm (bunch length) the collisions will be distributed over 300 ps (Fig. 1 left panel). Precise association of collision tracks or jets would help mitigate the pile-up. If this can be done for charged particles at high transverse momentum with particle tracking detectors this approach will be much more difficult in the forward-backward region and even impossible for neutral particles. In this case only TOF techniques can be applied as shown on the right panel of Fig. 1, where the two crossing bunches are symbolized by blue and red bars while their overlapping area is represented by a white bar. Events generated in the middle of the experiment $(z = 0)$ will generate tracks arriving at the same time in the forward and backward regions. On the other hand events generated at any time of-center of the bunch-overlapping region will exhibit a TOF difference for the tracks generated in the forward and backward regions, as shown on Fig. 1 $(t_2 - t_4, t_5 - t_7, t_8 - t_{10})$. A mitigation factor of one order of magnitude necessitates a TOF precision of at least 30 ps.

In the domain of medical imaging a new generation of TOF-PET scanners offers significant improvements in image quality. Commercially available TOF-PET have a time resolution ranging from 550 ps to 650 ps *FWHM* allowing to improve the reconstructed image signal-to noise ratio (S/N) by about a factor 2 as compared to standard PET scanners with non TOF capability and a time resolution of about 2 ns [3]. This corresponds to a spatial resolution of approximately 9 cm along the Line of Response (LOR), only sufficient to reduce the loss in image quality aggravated by the size of obese patients. Ongoing research work is aiming at a target value

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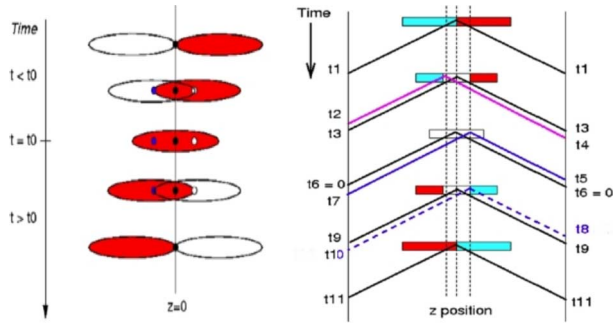

Fig. 1. Schematics of bunch crossing (left) and TOF (right) in the forward and backward directions of particles generated by events created in different positions of the overlap region.

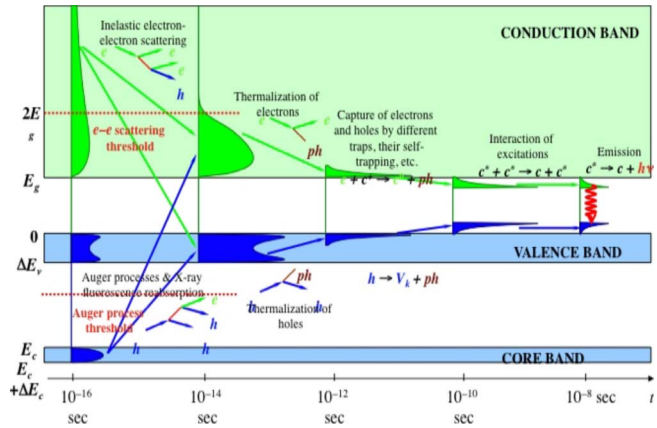

Fig. 2. Schematics of the relaxation mechanism time scale in a scintillator. (From A. Vasiliev [6]).

of 100 ps to 200 ps *FWHM* in coincidence time resolution (CTR) that would lead to a spatial precision of 1.5 cm to 3 cm along the LOR, allowing the localization of the organ under examination (e.g., prostate, pancreas or lymph nodes) and an efficient rejection of the background generated by other organs. This is the aim of the EndoTOFPET-US project, funded by the European Commission in the frame of the FP7 programme, for the development of new biomarkers for the pancreas and prostate cancers [4]. Despite being a significant improvement over standard PET cameras, this precision does not yet allow a direct 3-D reconstruction of a PET image, which is the ultimate goal. This however requires a CTR of about 10 ps for a spatial resolution of 1.5 mm along the LOR. Such a precision would allow an on-line image reconstruction of unprecedented S/N ratio eliminating the time consuming iterative or back-projection algorithms.

Hadron therapy would also greatly benefit from a fast on-line monitoring of the dose delivered during proton or carbon therapy treatment, requiring very high sensitivity, high resolution and fast reconstruction imaging of β^+ emitting isotopes produced by beam or target spallation processes during the irradiation [5].

II. Limits of Traditional Scintillation Mechanisms

The timing performance of a scintillator is directly related to the density of scintillation photons produced in a time frame corresponding to the targeted time resolution. A high light yield (LY) and a short decay time (τ_d) are mandatory as in first approximation the initial photon density is given by LY/τ_d. But this is an approximation only as scintillators are also characterized by a rise time (τ_r), which delays the emission of the first produced photons, increases their time jitter and reduces accordingly the time resolution of the scintillator.

The rise time results from a complex relaxation mechanism of the hot electron-hole pairs produced by the interaction of ionizing radiation with the scintillator crystal before the luminescent centers of the scintillator can be activated. This mechanism has been described in detail by one of us in [6] and is illustrated in Fig. 2.

The different steps can be summarized as follows:

1) 10^{-16} to 10^{-14} seconds: production of primary excitations by interaction of ionizing particles with the material. For very high incident particle energy, the excitations are essentially deep holes h created in inner core bands and hot electrons e in the conduction band. Then in a time scale of a few femtoseconds, a large number of secondary electronic excitations are produced through inelastic electron-electron scattering and Auger processes with creation of electrons in the conduction band and holes in core and valence bands.
2) 10^{-14} to 10^{-12} seconds: The multiplication of excitations is stopped when all electrons in the conduction band have an energy smaller than $2E_g$ (e-e scattering threshold) and all holes occupy the valence band if there is no core band lying above the Auger process threshold (general case). At this point starts the thermalization of electronic excitations with production of phonons, leading to low kinetic energy electrons in the bottom of the conduction band and of holes in the top of the valence band.
3) 10^{-12} to 10^{-10} seconds: This stage is characterized by the localization of the excitations through their interaction with stable defects and impurities of the material. For example, electrons and holes can be captured by different traps or self-trapped in the crystal lattice. Excitons, self-trapped excitons, self-trapped holes (V_K centers) can be formed with emission of phonons. Localization of excitations induces a local polarization of the crystal lattice (creation of polarons).
4) 10^{-10} to 10^{-8} seconds: The last step is related to migration of relaxed excitations and radiative and/or non-radiative recombination. It ends-up with the luminescence of emitting centers excited by the final electronic excitations (correlated electron-hole pairs, excitons, separated electrons and holes, etc.) through the sequential capture of charge carriers.

It clearly appears that the radiative transition on the activator ion or on the intrinsic luminescent center only takes place after a complex relaxation mechanism of the primary electron-hole pairs that can last several nanoseconds. The process being stochastics, large statistical fluctuations are therefore induced for the generation of the first scintillation photons at the origin of the observed rise time. There is therefore an intrinsic limit to the time resolution that can be achieved by a scintillator. It is

related to the time fluctuations in the relaxation process that can be estimated to be of the order of 100 ps.

If the required time resolution of the order of 10 ps mentioned in the introduction is to be achieved, a different approach would be needed. From this point of view an interesting phase of the relaxation mechanism is the thermalization step when the hot electrons and holes have passed the ionization threshold. The creation of acoustic and optical phonons in the lattice is the source of transient phenomena that could be exploited to obtain a time tag for the interaction of ionizing radiation with a precision in the picosecond range. Two such mechanisms, hot intra-band luminescence and transient absorption are described in Sections III–VI.

III. Requirements for Scintillator Intrinsic Time Resolution

The process of registration of the moment of the interaction of an ionizing particle with the scintillating crystal involves at least four stages: (1) interaction of ionizing particle with the medium with production of hot excitations in scintillator, (2) relaxation of hot excitation to the light emitting states and emission of light, (3) propagation of light to the detector, and (4) interaction of light with the photodetector. Each of these stages can be subdivided into individual sub-stages. The first process is responsible for the spatial fluctuations of the excited region produced by the ionizing particle. Taking into account a required overall time resolution of 30 ps we can conclude that spatial dimensions of the excited region should be less than 1 cm. This can be well satisfied in case of PET when energies of ionizing particles are low, but for HEP we have to optimize the geometry of the detector in order to bring the interaction region closer to the light detector. In any case, the overall detector response function is the convolution of the response functions of scintillator and light detector and propagation functions at the stages (1) and (3). Therefore the typical scintillator intrinsic time resolution should be better than 10 ps. The purpose of this paper is to concentrate on the light production mechanisms allowing achieving such a resolution, the other factors being considered in other reports [7], [8].

The important parameter for time resolution is the mean time of the registration of the first photon. If we concentrate on scintillator itself (assuming that all other stages are fast and have no losses of photons—no photon deletion) then for the model of a single exponential decay with characteristic decay time τ_d and rise time τ_r the mean time of registration of the first photon can be estimated as $t_{1st} = \tau_d/N_{ph}$ for $\tau_r < \tau_d/N_{ph}$ and $t_{1st} = \sqrt{2\tau_r\tau_d/N_{ph}}$ for the opposite case, where N_{ph} is the total number of emitted photons [8]. These estimations are obtained for a simple model of energy transfer in scintillators, when the rise of luminescence is characterized by single energy transfer step (which is not correct for most energy transfer processes in scintillators). In order to achieve small t_{1st} we have to reduce both τ_d and τ_r. In some applications with not too high rate (e.g., PET) we can use moderate τ_d and high value of N_{ph}. But if the rate of events is high (for HEP), the decay time should also be fast to avoid pile-up. We have therefore to consider some ultrafast events in solids such as hot luminescence and transient absorption.

IV. Hot Intra-Band Luminescence

The intra-band luminescence in solids is a weak intrinsic fluorescence which exists in all solids. It is the emission of photons during the second stage of the above-mentioned relaxation of electronic excitations. The theory of such hot luminescence has been developed more than 40 years ago. This kind of emission is very weak and was investigated experimentally mainly under electron beam excitation [9]–[15]. The energy of electrons was below the Cherenkov threshold (typical energy is about 100 to 300 keV). For higher electron energies this intra-band emission can be confused with Cherenkov radiation since its properties are similar to that of Cherenkov radiation (except for the angular dependence): the spectrum is wide, characteristic time is shorter than the electron pulse duration.

There are many different approaches, which describe the process of phonon-assisted relaxation of hot electronic excitations (see, e.g., [16]–[19]) based on different approximations. The simplest one is the quasi-classical description of relaxation as a set of kinetic equations with account for emission and absorption phonons and emission of photons. The first case to be considered is the case of uniform density of states (DOS).

A. Intra-Band Emission in Case of Uniform DOS

Intra-band luminescence is connected with the stage of relaxation, when electronic excitations are still not thermalized, but their energy is not enough to produce additional electronic excitations. Schematically the energy distribution of these electrons and holes is shown in Fig. 2 at 10^{-14} s. Therefore the kinetic energy of electrons and holes before this stage of relaxation is distributed between 0 and the forbidden gap energy E_g. Fig. 3 shows this region in some details. There are a lot of processes, which can occur with such excitations. The main process, which controls the thermalization of the excitation is the emission and absorption of phonons, namely longitudinal optical (LO) and acoustical (LA) phonons in ionic crystals. The rate of electron-phonon scattering was investigated in details in multiple papers. Recent estimations for scintillators can be found in [20]. Unfortunately these evaluations are correct only for parabolic energy dispersion law and can be used for real crystals only as rough estimations. The general trends are the following. The rate of LO phonon emission depends on the LO phonon energy $\hbar\Omega_{LO}$, the carrier effective mass and the so-called effective dielectric permittivity, which describes the response of ionic subsystems of the crystal to the electric field of the carrier. This rate is about 3×10^{14} s^{-1} for high $\hbar\Omega_{LO} = 100$ meV (fluoride and oxide crystals) and slightly lower for low $\hbar\Omega_{LO} = 10$ meV (CsI). Nevertheless, the rate of energy relaxation differs significantly. For a secondary electron with energy say 4 eV about 40 phonons should be emitted during energy relaxation, whereas in case of CsI the number of emitted phonons is about 400. Therefore the total time of energy relaxation from 4 eV to 0.1 eV is about 0.1 ps for 100 meV phonons and about few picoseconds for 10 meV phonons. These estimations were performed for constant value of the effective mass. In most crystals the effective mass of holes is larger than that of electrons.

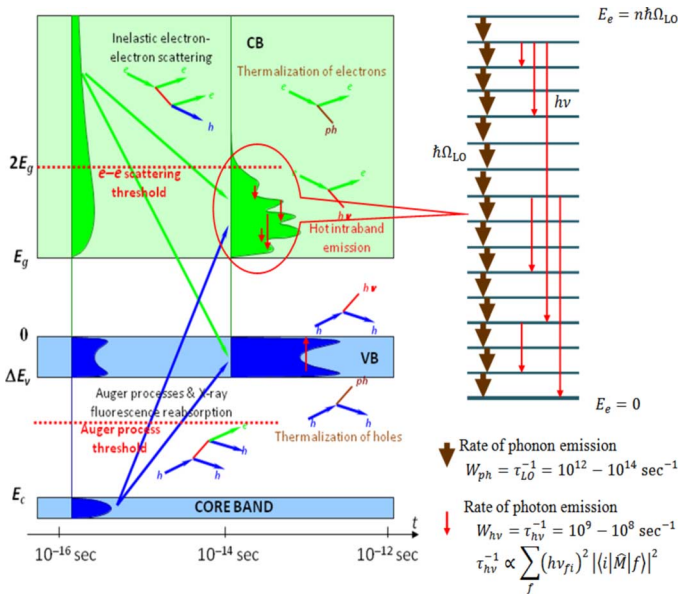

Fig. 3. Scheme of hot intra-band luminescence.

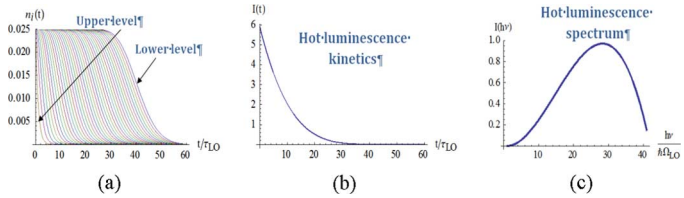

Fig. 4. Population of the different states (left panel), kinetics (middle panel) and spectral distribution (right panel) of intra-band luminescence in the case of uniform DOS and uniform initial population of all states from $E_e = 0$ to $E_e = 40\ \hbar\Omega_{LO}$.

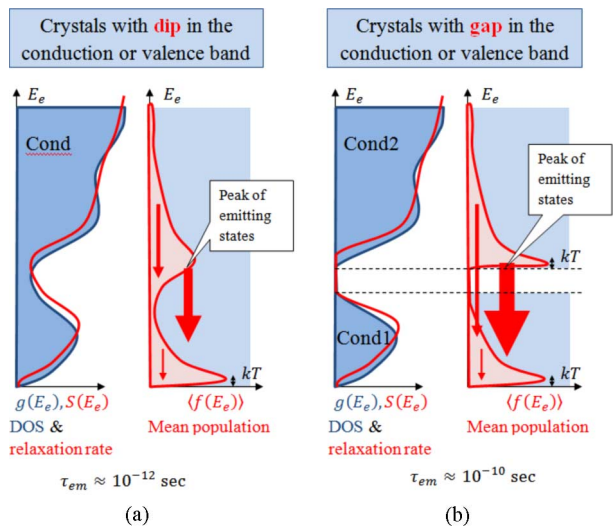

Fig. 5. Relaxation of electron states in crystals with dip (left panel) and gap (right panel) in the conduction band. Blue area shows the density of states $g_e(E_e)$, red line represents the rate of energy relaxation $S(E_e)$ due to interaction with phonons, pink area represent the mean population of the excited states $\langle f(E_e)\rangle$.

Therefore the relaxation of holes in the valence band is typically slower than the relaxation of electrons.

During this relaxation electrons and holes can emit photons. Carriers with kinetic energy E can transit to the state with lower kinetic energy E' with emission of photons with energy $\hbar\omega = E - E'$. This process is not allowed in case of parabolic band (it is well-known that free electrons cannot absorb or emit photons due to momentum conservation law). Therefore the band structure should have at least two branches in order to generate photons in 'vertical' transitions, and the increase of the branches number is preferable. Fortunately all crystals with relatively wide bandgap have many branches in the kinetic energy region up to E_g. The rate of photon emission is proportional to the square of $\hbar\omega$. The rate of dipole allowed radiative transition is about $10^8 - 10^9$ s^{-1}. Taking into account the time of relaxation with phonon emission one can estimate the yield of this hot intra-band luminescence as $10^{-5} - 10^{-3}$.

Fig. 4 demonstrates the main features of this emission. The decay of the emission is controlled by the lifetime of the population of the corresponding level. Its duration corresponds to the above-mentioned duration of energy relaxation (from 0.1 ps to few picoseconds). The decay is strongly non-exponential. The typical spectrum mainly depends on the band structure. For a uniform density of states it is represented by a smooth curve with an increase to the high-energy end of the spectrum. The upper energy of this spectrum corresponds to the threshold of inelastic scattering of electrons/holes with production of new electronic excitations, i.e., about the width of the forbidden gap E_g.

B. Intra-Band Emission in Case of Dips or Gaps in DOS

The analysis presented in the previous subsection shows that in case of structure-less DOS the yield of the intra-band luminescence is not sufficient for most applications. The number of carriers which are created by 511 keV photons in a crystal with $E_g = 5$ eV is about 5×10^4. Taking into account the estimated yield of this fast emission we can conclude that only in systems with low phonon frequencies the number of emitted photons can be higher than 10. Nevertheless, in crystals with a more complex distribution of density of states the situation should be different.

The rate of energy relaxation $S(E_e)$ of an electron with energy E_e depends on the phonon energy and the density of final states. For instance, in case of one LO phonon branch the rate $S(E_e) = \hbar\Omega_{LO}/\tau_{LO}$, and τ_{LO}^{-1} is proportional to the density of final states $g_e(E_e - \hbar\Omega_{LO})$. If this density has a dip (see Fig. 5, left panel), the energy relaxation rate should also have a dip (shifted to higher energies by the phonon energy $\hbar\Omega_{LO}$). This dip results in the bottleneck in the relaxation, thus producing the peak in average energy distribution above the dip. Such structures of conduction band DOS (or valence band DOS) result in accumulation of carriers above the gap, and radiative transitions can be more prominent. The estimations show that the lifetime of carriers above the gap could be about 10^{-12} s, and the yield of intra-band emission can achieve 10^{-3}, which is large enough to allow fast tagging of events for high energy particles. Moreover a proper engineering of the crystals could even open the way to TOF-PET systems, where the TOF information is provided by photons produced by this fast mechanisms whereas the energy resolution is obtained from the much more intense standard scintillation mechanism.

Even more interesting situations occur in crystals with a gap in the conduction or valence band. If the energy width of the gap is higher than $\hbar\Omega_{LO}$, the transition of carriers from the upper sub-band to the lower sub-band can be assisted only by multi-phonon emission, and the rate of this process is much lower than the rate of allowed one-phonon emission. If the energy width of the gap is about 1 eV, the lifetime of the states above the gap can achieve 10^{-10} s, and the carriers are partially thermalized at the bottom of the upper subband. Therefore the yield of the corresponding photon emission can be as high as 10^{-2}. Such gaps in the conduction band are often observed in band structure calculations of crystals with d-elements (for instance, CeF_3, $ZnWO_4$, etc.). Additional experimental investigation of such inter-sub-band luminescence is required. The advantages of this fast luminescence can be realized if the main (traditional) luminescence is quenched (and therefore no slow emission will be observed).

If the gap width is about a few electron-volts (this occurs mainly in the valence band), the conditions for cross-luminescence are realized. This case is well studied for many crystals, see e.g., [21].

V. Transient Absorption

An important factor when considering the interaction of high-energy charged particles with crystals is the significant size of the interaction region in space. The energy deposit in the crystal is due to ionization, bremsstrahlung and pair creation along the particle's trajectory. Moreover heavy relativistic particles lose a fraction of their energy through non-ionization loss processes (NIL). As an example a 1 GeV proton has a de Broglie wavelength of ~ 0.1 fm and can interact with individual nucleons in the crystal. The projectile shares its kinetic energy with the target nucleons by elastic collisions and a cascade of nucleon-nucleon collisions takes place. It creates a variety of atom vacancies and interstitials in a very short time along the projectile path. Moreover a relativistic particle moving with a speed $v \sim c$ in a crystal with a high refractive index (typically $n \sim 2$ in a majority of scintillators used in particle detectors) produces Cherenkov radiation. Since a relativistic particle energy greatly exceeds the Cherenkov radiation threshold the intensity of this radiation is likely to exceed the hot intra-band luminescence in a majority of crystals. As it will be difficult to discriminate the hot intra-band luminescence from the Cherenkov this can give rise to non-linearity and difficulties for precise energy measurements in high energy physics experiments.

An important property of the energy loss in the crystal by ionizing radiation is the polarization of the environment near the particle trajectory and the formation of hot polaronic states. When reaching an energy of less than $2E_g$ free electrons created by an incident particle interact strongly with lattice phonons and create free polaronic states. In addition to possible radiative transitions as explained in the previous section such transient states may cause electronic absorption transitions if the crystal is illuminated by an external light source. Thus, in addition to hot intra-band radiative transitions as shown in Fig. 5, absorption transitions can be observed as soon as the population of the lower levels of the conduction band exceeds the one of the upper levels, which usually takes place in less than 10^{-12} s. The observation of this absorption must be done before the capturing of the free carriers by luminescent centers and/or shallow traps. The time scale of this process is comparable to the scintillation rise time of a typical scintillation crystals doped with Ce^{3+} activator and does not exceed 100 ps. GEANT4 simulation shows that in a scintillator like LSO:Ce the energy deposit by 100 GeV electrons in the first 100 ps of interaction is ~ 520 MeV. Considering that the energy to create a free polaron is about $2E_g$ (i.e., about 12 eV for LSO) the maximum number of free polarons, which can be produced does not exceed $5 * 10^7$. They will provide a small and probably undetectable level of optical absorption.

On the other hand the polarization of the medium by the ionizing incident particle provides a better sensibility for the particle detection by optical absorption as it involves the local distortion of the crystal lattice on a relatively large volume. The ionization of atoms in the lattice produces local charge imbalance. That leads to a short-term restructuring of the lattice for charge compensation. For materials containing tightly packed cells on the basis of, for instance W, Mo, Al, Si oxide polyhedra, the charge imbalance induces a distortion of these polyhedra. In the case of energy deposit by heavy relativistic particles the formation of atom vacancies and interstitials leads to the instantaneous perturbation of the crystal field and to the formation of distorted polyhedra. It must be noticed that the process of Cherenkov radiation is the direct consequence of the polarization of the medium along the particle path. An interesting case is the one of crystals with the bottom of the conduction band formed by **nd** levels of atoms of the lattice, such as tungstate or lutetium-based crystals. The distortion of the polyhedra will inevitably lead to a lowering of their symmetry and, as a consequence, to the creation of low-symmetry crystal field components and to additional splitting of **d** orbitals. Consequently, the absorption near the fundamental absorption edge can be temporarily modified and detected by short laser pulses of appropriate wavelength with a duration of 5–10 ps and a repetition rate of 100 GHz. Such techniques are widely used in the study of ultra-fast phenomena in optical media [22]. With a proper choice of the medium the modulation of an external high repetition rate optical carrier should allow a precise time-tagging at the level of 10–20 ps following the principle of heterodyne detection already described by one of us [23].

VI. Conclusions

The demand is increasing for fast scintillators to answer the requirements of fast time tagging and pile-up mitigation in HEP detectors at future high luminosity colliders as well as for image S/N improvement and millimetric direct 3D acquisition in PET scanners. It is unlikely that the challenge of 10 ps time resolution can be reached on the basis of standard scintillation process, which occurs after a complex and much longer than 10 ps relaxation mechanism of the primary electron-hole pairs.

On the other hand transient phenomena taking place during the thermalization of electrons and holes when their energy has dropped below the multiplication threshold can be exploited to obtain a time tag in the picosecond range. Two such phenomena are described in this paper, hot intra-band luminescence and

transient absorption, both resulting from electron and/or hole coupling to the lattice acoustic and optical phonons. For crystals where the density of states is non-uniform in the conduction band sub-10 ps radiative transitions or absorption states could be observed between sub-bands.

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