Non-Linear Optical Phenomena in Detecting Materials as a Possibility for Fast Timing in Detectors of Ionizing Radiation

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***Abstract*— The time resolution of the detectors currently in use is limited by 50–70 ps due to the spontaneous processes involved in the development of the response signal, which forms after the relaxation of carriers generated during the interaction. In this study, we investigate the feasibility of exploiting sub-picosecond phenomena occurring after the interaction of scintillator material with ionizing radiation by probing the material with ultra-short laser pulses. One of the phenomena is the elastic polarization due to the local lattice distortion caused by the displacement of electrons and holes generated by ionization. The key feature of the elastic polarization is its short response time, which makes it prospective for using as an optically detectable time mark. The nonlinear optical absorption of femtosecond light pulses of appropriate wavelength is demonstrated to be a prospective tool to form the mark. This study was aimed at searching for inorganic crystalline media combining scintillation properties and non-linear absorption of ultra-short laser pulses. The nonlinear pump-and-probe optical absorption technique with 200 fs laser pulses was used to study the effects in lead tungstate, garnet-type, and diamond scintillator crystals.**

***Index Terms*— Fast timing, inorganic scintillation material, particle colliders, two–photon absorption.**

I. Introduction

THE upcoming experiments in high-energy physics require the time resolution of scintillation detectors better than 10-20 ps. This is not feasible using the conventional scintillation detectors with the time response limited by the time of carrier relaxation and transfer to the radiative recombination centers in the scintillator material.

In this study, we investigate the feasibility of using the phenomena occurring in parallel with the carrier relaxation within the very first picoseconds after the ionization starts. One of the phenomena is the elastic polarization due to the local lattice distortion caused by the displacements of electrons and holes generated by the ionization. This local distortion in the lattice results in redistribution of the density of states (DOS) of electron in the conduction band in close vicinity of the hole. The key feature of the elastic polarization is its short response time, which makes it prospective for using as an optically detectable time mark. Nonlinear optical absorption of femtosecond light pulses at appropriate wavelength is considered to be a tool to form the mark.

The effect of elastic polarization should be observed in many crystalline compounds. According to our estimations, the strongest effect should be observed in compounds with the bottom of the conduction band formed by $n\mathbf{d}$ orbitals of the lattice cations. According to the crystal field theory, these orbitals are most sensitive to distortions of the crystal field in the vicinity of emitting centers. Thus, the crystals with lattice cations having strong contribution of **d** orbitals in conduction band (tungstates, molybdates, rare-earth and yttrium garnets, perovskites, oxy-orthosilicates, etc.) might be good candidates for using them as timing tools.

Our study was aimed at searching for inorganic crystalline media exhibiting good scintillation properties as well as strong non-linear absorption of ultra-short laser pulses. Accordingly, we selected three scintillation crystals: self-activated lead tungstate $PbWO_4$ and mixed $Gd_3(Ga_{0.5}$-$Al_{0.5})_5O_{12}$:Ce and yttrium $Y_3Al_5O_{12}$:Ce garnets. Lead tungstate is currently the most extensively used scintillation material in high energy physics experiments [1], while the recently developed mixed garnet crystals showed fast response and highlight yield up to 56000 ph/MeV [2]. YAG:Ce is also fast and bright scintillation material exhibiting one of the highest radiation hardness among the scintillators currently in use under both gamma and high energy proton irradiation [3]. Recently, we confirmed the influence of ionization at 122 keV on the two photon absorption of femtosecond laser pulses in $PbWO_4$ crystals [4]. The observed effect encouraged our further study in this direction to develop a novel detecting technique exploiting the interaction of short laser pulses with crystalline media excited

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by ionizing radiation. The results of this study are presented in the current paper. In addition to pure scintillation materials synthetic diamond was probed with two-photon absorption.

II. One and Two-Photon Absorption in Scintillation Materials

One-photon absorption is extensively used to monitor different effects in ionizing radiation detectors. For instance, nanosecond laser pulses are used to monitor radiation damage effects in PWO crystals [5]. In fact, this technique enables monitoring slow change in the detector material properties, particularly accumulation of the color centers under ionizing radiation.

One-photon absorption is not convenient to explore changes in the DOS due to strong absorption of single photons via electronic transitions between valence and conduction bands. This is due to the origin of the bands of the majority of inorganic wide band gap compounds: p electronic states form the top of valence band, whereas d and f states of metal ions dominate in forming the bottom of the conduction band. Dipole-allowed p-d transitions result in the absorption coefficient for the interband transitions at the order of 10^5 cm^{-1}.

The selection rules for two-photon absorption are different [6]: p-d transitions become forbidden and their rate falls by orders of magnitude. Consequently, the photons absorbed via two-photon absorption propagate relatively long distances in the crystal.

Two-photon absorption can involve photons of the same frequency generated by the same laser or simultaneously available photons of different frequencies [7]. The two-photon absorption involving one pump and one probe photon is a convenient tool for studying both time and spectral parameters of the interband absorption. Recently, the pump-probe technique was exploited to study $PbWO_4$ crystals [8]. The pump-induced changes in material properties resulting in modified probe absorption in the sample volume where the pump and probe beams spatially overlap have been recorded.

III. Experimental

Two pump and probe setups were exploited in our study as described in [8]. In the first setup, the 140 fs long pulses of the second harmonic of Al_2O_3:Ti^{3+} laser radiation was used to generate both the pump beam with a fixed wavelength of 395 nm and a white continuum ranging 400 to 1100 nm, which was generated in water by a fraction of the fundamental laser radiation at 790 nm and used as a probe.

The second setup was used for pump and probe measurements at different pump beam wavelengths from 346 to 650 nm. The experiments were performed at different pump pulse energies. The system was based on a femtosecond Yb:KGW laser producing 200 fs pulses at 1030 nm with repetition rate of 30 kHz.

The samples of $PbWO_4$ crystals under study were fabricated at the Bogoroditsk Technical Chemical Plant (Russia) for PANDA experiment at FAIR (Darmstadt, Germany). The garnet samples were produced by ISMA (Kharkov, Ukraine). All the samples under study had a thickness of 1cm.

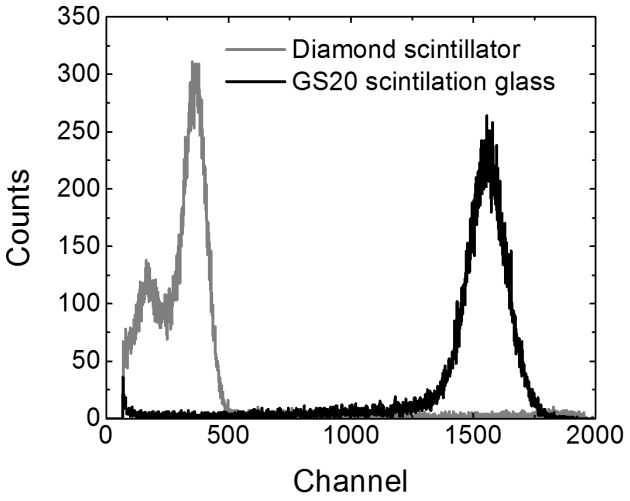

Fig. 1. ^{239}Pu alpha-particle amplitude spectra measured with diamond scintillator #11 (dots) and light GS20 scintillation glass (crosses).

The diamond samples were produced by (ADAMAS-BSU), Minsk, Belarus, using the High Pressure High Temperature (HPHT) technique. The samples had yellow coloration due to the presence of nitrogen. The cutoff of the transmission spectrum of both samples was found to be near 420 nm. They had dimensions of 4x4x0.3 mm. Both the samples under study had the charge collecting distance of $\sim$ 0.1 mm and were tested to detect the ionizing radiation via electrical signal readout with the electrodes evaporated on their larger surfaces. Both the crystals exhibited a luminescence band peaked in the vicinity of 530 nm and having a broad excitation band peaked at $\sim$340 nm. One of the samples (4.8) was not scintillating, however another one (11) had a clear scintillation effect under excitation by alpha-particles. The scintillating crystal had a photo-excited luminescence intensity by one order of magnitude larger. In addition to HPHT diamonds a clear diamond produced by Chemical Vapor Deposition (CVD) method with dimensions 4x2x0.15 mm also was studied. Figure 1 shows the room temperature amplitude spectra of ^{238}Pu alpha-particles measured with 0.3 mm thick diamond crystal and a reference spectrum obtained with light scintillation glass GS20. CWD diamond also showed scintillations at the detection of alpha-particles, however its light output was found to be three times smaller than of HPHT diamond.

Luminescence of synthetic diamonds is well described [9], [10] and is related to the defects, which appear in the crystals due to peculiarities of technology. The green luminescence is caused by complex defects containing nickel and nitrogen.

All the measurements were performed at room temperature.

IV. Two-Photon Absorption in $PbWO_4$ Single Crystal

The change in optical density for the probe beam induced by the pump at the wavelength of 394 nm is presented as a function of the delay between pump and probe in Figure 2. The spectral dependence of the two-photon absorption is described elsewhere [4], [8]. The nonlinear response has a

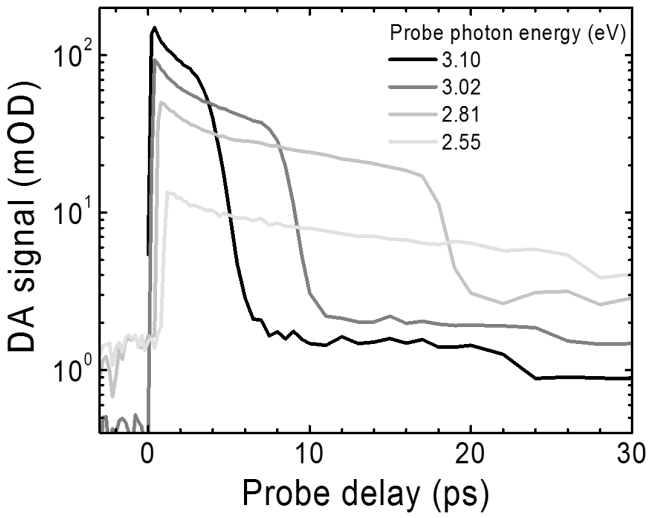

Fig. 2. Kinetics of differential absorption in PWO scintillation crystal for 394 nm pump at different probe wavelengths (indicated).

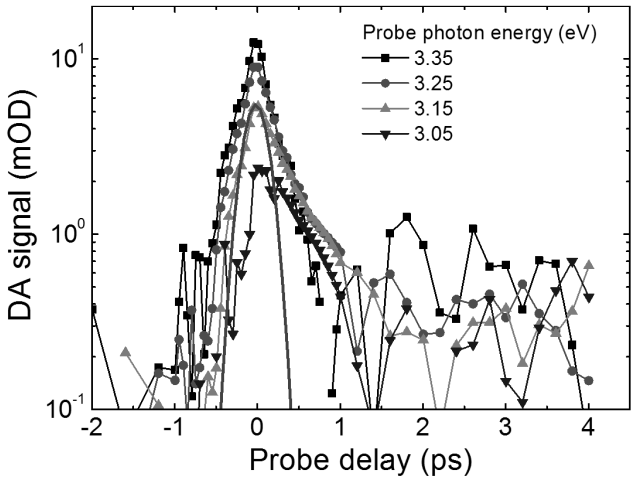

Fig. 3. Kinetics of differential absorption in $Gd_3(Ga_{0.5}\text{-}Al_{0.5})_5O_{12}$:Ce scintillation crystal for 360 nm (3.44 eV) pump at different probe energies (indicated).

short rise time, decays with characteristic time constant of few picoseconds, and a slower decay afterwards. The fast decrease of the induced absorption at the rear edge of the response is explained by the separation of the pump and probe beams in space, since the beams are focused on the same spot of the sample surface at slightly different angles. The duration of the nonlinear absorption depends also on the pump and probe wavelengths due to the dispersion of the light velocity.

The most important feature of the response is that the leading edge of the differential absorption is limited by laser pulse shape rather than by material properties. Thus, the processes responsible for the nonlinear absorption occur on femtosecond domain.

V. Two-Photon Absorption in $Gd_3(Al_{0.5}\text{-}Ga_{0.5})_5O_{12}$:Ce Single Crystal

Figure 3 shows the change in optical density at several wavelength induced by the pump at the wavelength of 360 nm (3.44 eV) in $Gd_3(Ga_{0.5}\text{-}Al_{0.5})_5O_{12}$:Ce (GAGG:Ce) crystal. GAGG:Ce has more complicated band structure than $PbWO_4$. The crystal contains a narrow sub-band formed by Gd^{3+}ions bellow the conduction band in the band gap, which equals 6.4 eV in this material. Moreover, the crystal is doped with Ce^{3+} ions causing two absorption bands peaked in the vicinity of 340 and 440 nm. Therefore, pumping of the sample was carried out by pulses at the wavelength between the two absorption bands of activator.

Note that the two-photon absorption in GAGG:Ce is an order of magnitude weaker than that in $PbWO_4$. Figure 4 shows the spectral dependence of the two-photon absorption at different delays after the pump pulse at 360 nm.

The two–photon absorption peak is observed when the total energy of pump and probe exceeds the band gap. As evident in Fig. 4, the effect appears, when the energy of the probe photon exceeds 3 eV.

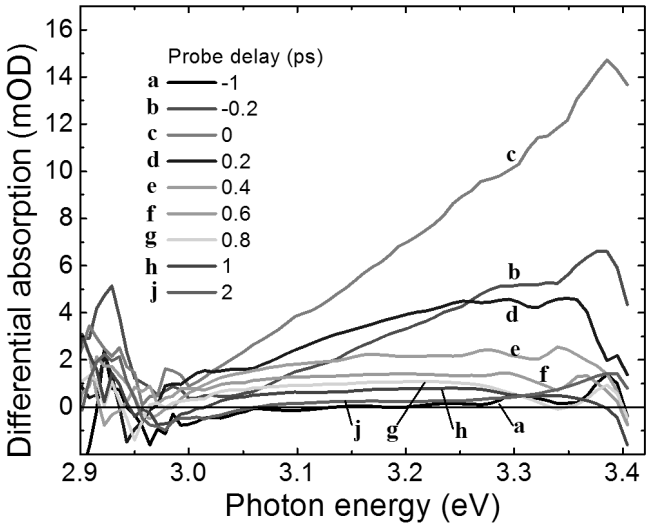

Fig. 4. Spectrum of differential absorption of $Gd_3(Ga_{0.5}\text{-}Al_{0.5})_5O_{12}$:Ce scintillation crystal for 360 nm (3.44 eV) pump at different delays between pump and probe (indicated).

VI. Two Photon Absorption in $Y_3\ Al_5\ O_{12}$:Ce Single Crystal

The band gap of yttrium aluminum garnet is close to that of GAGG band gap, however, in contrast to PWO and GAGG, the DOS of conduction band in YAG is formed predominantly by 4d orbitals. Nevertheless, we did not observe two-photon absorption in YAG:Ce for the pump plus probe photon energies in the range 6.2-6.8 eV (see Figure 5). The drop in the spectrum in the vicinity of 6.9 eV is caused by contribution of scattered pump beam when the energies of pump and probe coincide.

When pump energy corresponds to the excitation to a higher level of d configuration of Ce^{3+} ions, the intracenter relaxation is observed in the kinetics of the differential absorption. Figure 6 shows this behavior at pump at 264nm (4.73 eV) and two different probe wavelength of 430 nm (2.9eV) and 700 nm (1.75 eV).

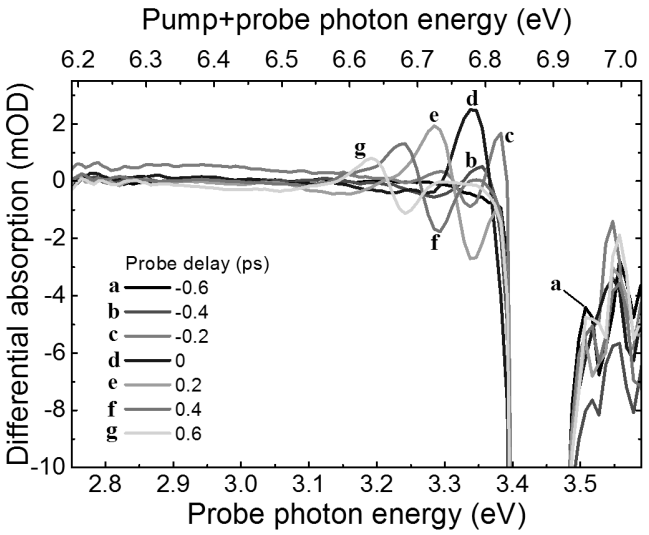

Fig. 5. Spectrum of differential absorption of $Y_3Al_5O_{12}$:Ce for 360 nm (3.44 eV) pump at different delays between pump and probe (indicated).

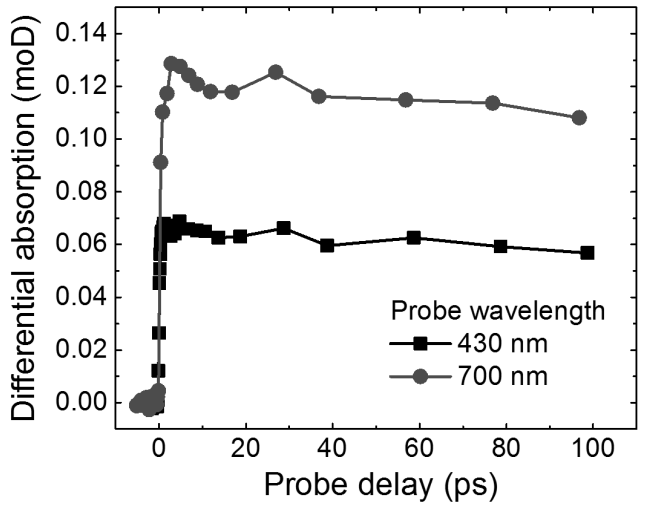

Fig. 6. Kinetics of differential absorption in $Y_3Al_5O_{12}$:Ce for 264 nm (4.73 eV) pump and two different probe wavelength.

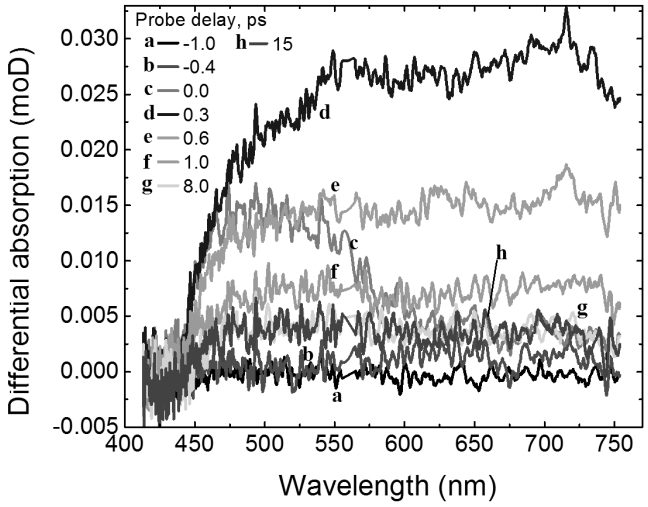

Fig. 7. Spectrum of differential absorption of diamond sample 4.8 at 395 nm (3.16 eV) pump at different delays between pump and probe (indicated).

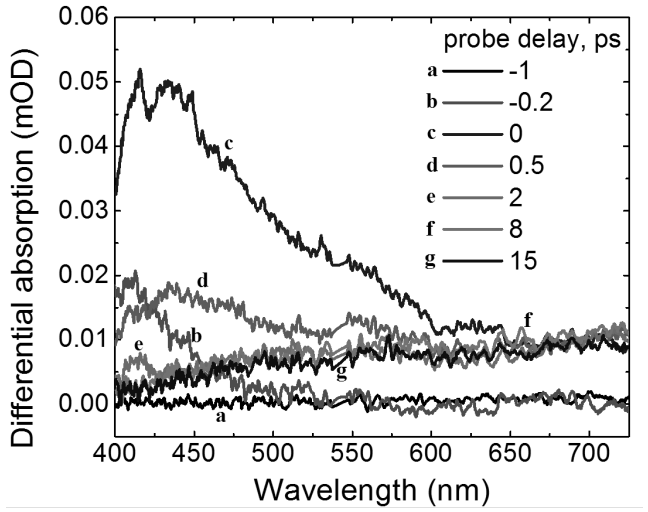

Fig. 8. Spectrum of differential absorption of CVD diamond sample at 264 nm (4.73 eV) pump at different delays between pump and probe (indicated).

Photons with energy 4.73 eV excite Ce^{3+} centers, the excitation promptly relax to the emitting state, while the probe photons throws the electrons up to conduction band. The observed decrease of induced absorption in time is caused by radiative recombination from the Ce^{3+} emitting level.

VII. Two-Photon Absorption in Synthetic Diamond Crystal

In contrary to the complex oxide crystals based on oxy-anionic complexes, diamond is a wide band gap indirect semiconductor having a relatively simple electronic band structure due to its monatomic network. Two-photon absorption in nitrogen-free diamond is studied in detail [11]. The lowest two-photon absorption band was observed to peak at the total energy of the two photons of 5.5 eV. This energy corresponds to the indirect transition to conduction band. The direct two-photon absorption was found to occur at 6.5 eV. We observed two-photon absorption in diamond samples under study. Figure 7 shows spectrum of differential absorption of diamond sample 4.8 at 395 nm (3.16 eV) pump at different delays between pump and probe.

The band peaked at$\sim$ 465 nm (2.6 eV) correlates fairly well with the energy of the indirect transition to conduction band. Another bands observed in the samples were peaked at $\sim$550 and 700 nm.

On the contrary, CVD diamond did not show similar spectra of differential absorption at 395nm pump. Strong differential absorption was found only at pump 264 nm as seen from Fig. 8. Contrary to sample 4.8 we observed strong band in blue range peaked at 430 nm and also two weak bands roughly peaked at $\sim$550 and 700 nm. So one can state that two last bands connected with defects which concentration is considerably larger in HPHT diamonds.

All bands of differential absorption in HPHT sample decay fast within the first picosecond and a slow decay component is

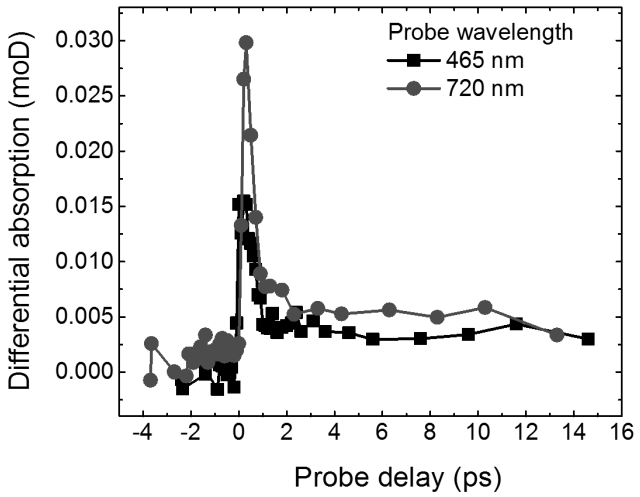

Fig. 9. Kinetics of differential absorption of HPHT diamond sample 4.8 at 395 nm (3.16 eV) pump and two probe wavelengths: 465 nm (2.7 eV) and 720 nm (1.74 eV).

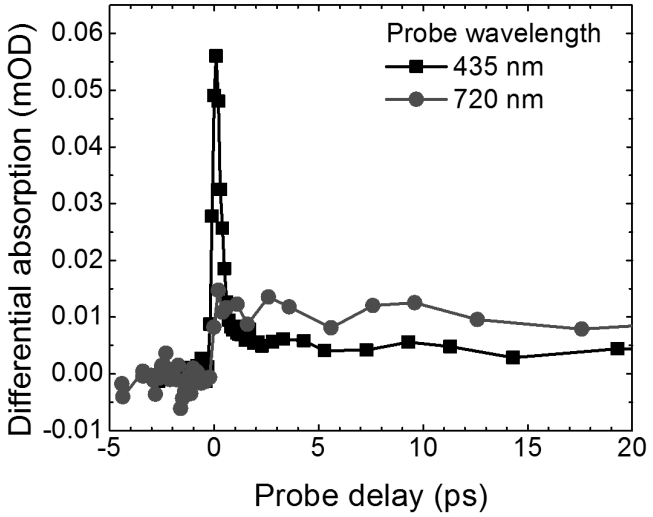

Fig. 10. Kinetics of differential absorption of CVD diamond sample at 264 nm (4.73 eV) pump and two probe wavelengths: 435 nm (2.9 eV) and 720 nm (1.74 eV).

practically absent (see Figure 9). The total energy of pump and probe photons for the 700 nm band was considerably smaller than band gap of diamond, so we concluded that defect centers are responsible for the two-photon absorption in this spectral region.

The different fast process of decay is observed in CVD diamond at UV pump and probing at 435 and 720 nm as seen from Figure 10. Due to small concentration of defects the signal is weak when probed with 720 nm. Also, contrary to colored HPHT diamonds, CVD diamond showed strong fast absorption at total energy 7.6 eV which correlates with data of [11]. The observed two-photon absorption effects in diamond demonstrate good perspectives of exploiting two photon and defect-related absorption for timing in radiation detectors.

When scintillating HPHT diamond sample is probed at 700 nm, the kinetics of differential absorption completely

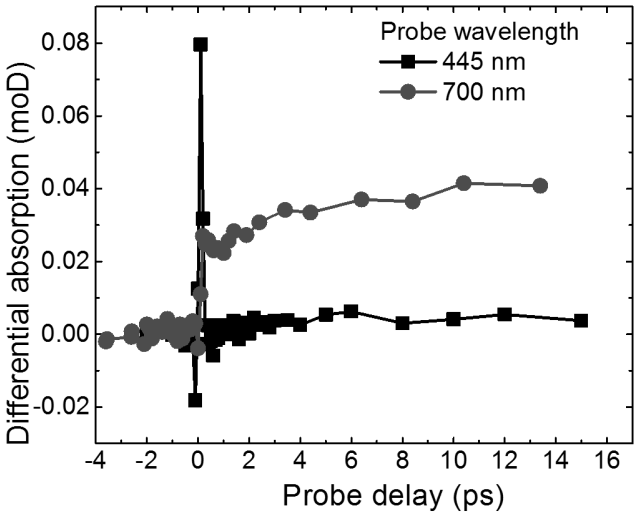

Fig. 11. Kinetics of differential absorption of scintillating diamond sample 11 at 395 nm (3.16 eV) pump and two probe wavelengths: 465 nm (2.7 eV) and 700 nm (1.7 eV).

changes form as seen from Figure 11. There are two rise process: fast, less than one ps, which is dedicated to direct excitation of defect and slow, a few ps, which to our mind is due to a population of the defect by capturing of the carriers from conduction zone.

The last process may be interesting to detect start of ionization in diamond at its interaction with ionizing radiation. In this case only probing of the diamond with near IR short laser pulses will follows to ionization pulse.

VIII. Outline of Detection Technique

The effects observed in our study might be exploited for timing of the interaction with ionizing radiation in parallel with the detection of scintillation signal in the same material. The change in two-photon absorption can be used to form a time mark to detect the initial moment of the interaction of the ionizing radiation with the detector material, while the scintillation signal provides the information on absorbed energy. The feasibility of this approach is not clear for Ce^{3+} materials, where the two-photon interband absorption is relatively weak. Meanwhile, in PWO the effect is quite strong and sensitive to the presence of ionizing radiation. Thus, the two photon absorption in this scintillator can be exploited to form the time mark. It is worth noting that in real applications based on the two-photon absorption there will be no need to use the sophisticated setups we use to optimize the detection system. The laser sources based on Nd^{3+}-doped crystals are commercially available at relatively low cost. The second harmonic of their radiation can be used to produce the light in the wavelength range of 500-530 nm, which is optimal for timing.

The light propagating along the scintillation crystal and reflected from the front face of the crystal could be used to observe the two-photon absorption. This configuration would also diminish the problems of spatial overlap of the pump and probe beams. The detection could be accomplished by using

a set of sub-picosecond pulses, e.g., 100 pulses with the total set duration of 0.1-1 ns. The set is injected into the crystal approximately at the time when the products of interaction in collider are expected to reach the detector. The initial moment of the interaction between the collision products and the detector material is detected by comparing the set of pulses reflected back out of the crystal and a reference set. In this scheme, the time resolution would be equal to the half of the time span between the pulses in the set. Other schemes are also feasible and should be worked out after a proper choice of the nonlinear optical response to be used.

Effects observed in diamond also suggest that start of ionization process in the material can be defined either by two-photon absorption or probing of population of the intrinsic defects.

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