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SPORT: A new sub-nanosecond time-resolved instrument to study swift heavy ion-beam induced luminescence – Application to luminescence degradation of a fast plastic scintillator

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ABSTRACT

We developed a new sub-nanosecond time-resolved instrument to study the dynamics of UV–visible luminescence under high stopping power heavy ion irradiation. We applied our instrument, called SPORT, on a fast plastic scintillator (BC-400) irradiated with 27-MeV Ar ions having high mean electronic stopping power of 2.6 MeV/μm. As a consequence of increasing permanent radiation damages with increasing ion fluence, our investigations reveal a degradation of scintillation intensity together with, thanks to the time-resolved measurement, a decrease in the decay constant of the scintillator. This combination indicates that luminescence degradation processes by both dynamic and static quenching, the latter mechanism being predominant. Under such high density excitation, the scintillation deterioration of BC-400 is significantly enhanced compared to that observed in previous investigations, mainly performed using light ions. The observed non-linear behaviour implies that the dose at which luminescence starts deteriorating is not independent on particles' stopping power, thus illustrating that the radiation hardness of plastic scintillators can be strongly weakened under high excitation density in heavy ion environments.

1. Introduction

Scintillators play a central role in various fields including particle detection and discrimination, which is in constant evolution, or in emerging fields such as hadrontherapy, where heavy particles are involved. The knowledge of the yields and lifetimes as well as the radiation hardness of scintillators when, instead of electrons or X-rays, high excitation density particles are used is essential. Time-resolved measurement is a major asset to investigate the excitation and energy transfer processes involved in luminescence, which often occur over a few nanoseconds at most. To our knowledge, the only device allowing for the investigations of scintillators dynamics under swift heavy ion irradiation was developed by Kazuie Kimura and collaborators [1–7]. With a resolution of about 85 ps, their instrument extended the field of investigations towards the primary processes under extremely high-density excitation. Especially, one of their remarkable outcome was to evidence a novel ultra-fast luminescence (UFL) in almost all of the insulators investigated [4–7]. UFL is a wide wavelength range emission that can be as short as several tens of picoseconds. This phenomenon cannot be attributed to known excited species but was interpreted to result from the relaxation of dense electron-hole plasma in the track core of swift heavy ions [4–7]. This is an example of the uniqueness of the phenomena induced by swift heavy ion irradiation, where energy deposition can be orders of magnitude higher than by electron or X-ray irradiation.

We present here the first version of a new device to study the dynamics of ion-beam induced luminescence (IBIL) using high stopping power heavy ion irradiation. Our instrument, called SPORT (French acronym for time-resolved optical spectroscopy), will allow for simultaneous UV–visible spectral and sub-nanosecond time-resolved measurements in a wide range of temperatures, thus enabling the discrimination of the different processes involved in scintillation and their evolution with irradiation. Our first investigations using SPORT were performed on a widely used plastic scintillator: BC-400, also known as NE-102A. Plastic scintillators cumulate many advantages such as fast and short time responses, facilitated shaping and low costs. Therefore they have been used for decades in many radiation applications [8]. Their main drawback is however the degradation of their luminescence properties under irradiation. It has long been known that the energy deposition in organic scintillators not only excites the fluorophores but

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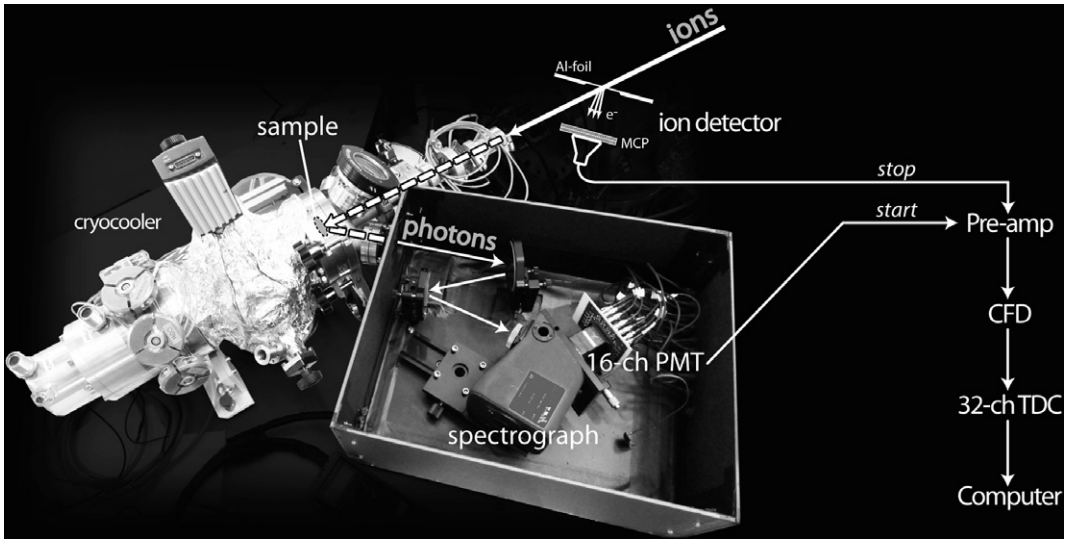

Fig. 1. SPORT instrumental set-up. Note that the second optical table, which extends the wavelength range with 16 additional channels, is absent on the picture since not used in this study.

also breaks chemical bonds, leading to luminescence quenching [9]. Accordingly, the previous IBIL studies of BC-400 revealed a degradation of scintillation as a function of irradiation dose [10–13]. However, these studies used light ions and were not time-resolved, as usually for studies of IBIL intensity degradation of plastic scintillators. We report here the first time-resolved IBIL investigations of BC-400 using swift heavy ion irradiation. Our work, focused on the dynamics of the scintillator and its evolution with ion fluence, brings new insights on the quenching type and the weakening of radiation hardness under high excitation.

2. Experimental

The instrumental set-up of SPORT is illustrated in Fig. 1. Basically, the principle of a time-resolved measurement with this instrument is to count photon in coincidence with ion hitting. Target samples are mounted in a UHV chamber on a cold finger tip (down to $\sim 10^{-9}$ mbar and 7.5 K). The sample holder is oriented normal to the ion beam and opens a 10-mm diameter window on the target. Photons are collected at 45° through two UV–vis windows and focused via spherical and planar mirrors on the entrance slits of two spectrographs (CP140-1604 and -1605 from HORIBA). They are finally counted using two 16-channel photomultiplier tubes (PMTs) disposed at the focal planes of the spectrographs (H10515B-04 and -20 from HAMAMATSU). This set-up allows to cover the 190–780 nm wavelength range with an average dispersion of ~14 nm/ch. Ions are detected with a home-made detector. Ions are passing through a 0.8 μm Al foil and the secondary electrons are detected by a tandem microchannel plate (MCP) (Fig. 1). After pre-amplification and constant fraction discrimination, the time-arrivals of the pulses from the PMTs and the ion detector are coded by a 32-channel multi-hit time-to-digital converter (TDC), which time resolution is adjustable down to 25 ps (V1290 from CAEN). Events (ion-photon coincidences) are sent to a computer and stored line by line, allowing for offline processing of the data. A detailed description of the final version of SPORT will be published elsewhere.

The sample investigated presently is a few mm-thick sheet of BC-400 from St-Gobain (equivalent to NE-102A). This ternary scintillator is composed of a polyvinyltoluene (PVT) matrix containing two fluorophores: *p*-Terphenyl (*p*-T) and 1,4-bis(5-phenyloxazol-2-yl) benzene (POPOP) [14]. The energy deposited by ionizing particles is transferred non-radiatively to *p*-T and then radiatively to POPOP. BC-400 has a maximum light output at 423 nm, which corresponds to the emission band of POPOP, and has rise and decay times of 0.9 and 2.4 ns respectively [14,15]. We set up SPORT in order to focus on the dynamics of this band and its evolution with ion fluence. Experiments were performed at the IRRSUD beam line of the Grand Accélérateur National d'Ions Lourds (GANIL) facility in Caen, France. The sample was irradiated at room temperature with 27-MeV $^{36}Ar^{10+}$ ions obtained from the injector cyclotron of GANIL facility operating in packages with a repetition rate of 13.45 MHz. According to SRIM code [16], these ions have a range of 11.3 μm and a mean electronic stopping power $\langle (dE/dx)_e \rangle$ of 2.6 MeV/μm. This value is by far the highest among the previous IBIL studies of BC-400 [10–13]. The ion beam was collimated between 16×16 mm^2 slits onto the sample at a mean fluence rate of 3×10^6 ion cm^{-2} s^{-1}. We used only one spectrograph and we present results on one channel of the PMT at 423 nm. Pulses from the PMT and the ion detector provided start and stop signals, respectively, on two channels of the TDC. The time resolution of the TDC was set to ~100 ps. The time-dependent emission curves of BC-400 were constructed by subtracting the time arrival of the photons to that of the ions and summing over the events of successive steps in fluence. As additional way to evaluate the time resolution of the instrument, and thanks to the multi-hit recording of the TDC, an ion self-correlation curve was constructed by subtracting the time arrival of consecutive stop signals, i.e. from the TDC channel provided by the ion detector.

3. Results and discussion

The ion self-correlation curve at final fluence, which represents the time spread between ions from successive packages of the cyclotron, is illustrated in Fig. 2. It is well described by a Gaussian function with a 0.6 ns-FWHM. The variance of this time spread represents twice the variance on the transit time from ion arrival to TDC time coding via ion detection and electronics. Therefore, any of the time jitters from this transit, for instance that of electronics, has to be less than $0.6/\sqrt{2} \approx 0.4$ ns.

The emission curve of the 423 nm band of BC-400 with 27-MeV Ar irradiation at final fluence is illustrated in Fig. 3. The rise part of the curve, defined from 10% to 90% of pulse height, spreads over 1.3 ns, which is higher than the 0.9 ns usually reported for this scintillator [15]. Assuming that this difference completely originates from time jitter, we can estimate the upper bound for the