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Analysis of linear energy transfer effects on the scintillation properties of $Bi_4Ge_3O_{12}$ crystals

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ABSTRACT

We analyzed the linear energy transfer (LET; energy deposited onto the target per unit length) effects on the scintillation properties of $Bi_4Ge_3O_{12}$ (BGO) with an emphasis on the dynamical aspect. We irradiated BGO with 20 MeV $H^{\pm}$, 50 MeV $He^{\pm}$, and 220 MeV C^{5+}. We observed that the rise and the decay of the scintillation temporal profiles are faster at higher LET. The faster decay at higher LET is attributed to the competition between the radiative transition of self-trapped excitons (STEs) localized at Bi^{3+} ions and the quenching caused by the interaction between STEs. The faster rise can be explained in terms of the competition between the quenching caused by the interaction between excited states and the formation of the STEs.

1. Introduction

Dense electronic excitation along the paths of energetic and heavy charged particles in solids has been extensively studied. At high density excitation, the interactions between the closely-located excited states often lead to the formation of columnar defects, known as ion tracks [1]. Several models have been proposed to describe the basic processes involved in the interaction between the excited states and their relaxation, which lead to permanent defect formation. Experimentally, the track formation threshold of the excitation density (linear energy transfer, LET; energy deposited onto the target per unit length) has been correlated with various material properties [1]. In addition to the studies of track formation, which is the final result of dense electronic excitation, the dynamics of the dense electronic excited states have been analyzed mainly by measuring luminescence or scintillation.

The dynamics of the excited states in insulators have been analyzed by Kimura et al. using time-resolved luminescence spectroscopy [2–7]. They examined the dynamical aspects of the excited state interactions including self-trapped excitons (STEs) [2] and core holes [4]. In addition, they found a fast and broad luminescence band in alkali halide crystals, which is attributed to an interaction similar to electron-hole plasma in semiconductors [6,7]. Our group has also analyzed the interaction of the excited states using luminescence spectroscopy in semiconductors [8,9] and α-Al_2O_3 [10]. Other groups also have reported the time-resolved luminescence from polystyrene [11], liquid scintillators [12], and naphthalene [13].

Recently, we began investigating the excited state dynamics in scintillators. Many inorganic scintillators are comprised of an insulator host and impurity ions as luminescence centers. In such systems, the energy of ionizing radiation is initially deposited mainly at the host, and a subsequent energy transfer occurs from the host to the luminescence centers. Thus far, we have shown the LET effects on the energy transfer process, i.e., the dependence of the rise in the scintillation temporal profiles on LET [14–16]. These LET effects are attributed to a competition between the quenching caused by the interaction of the excited states, and the energy transfer.

In this study, we focused on a scintillator in practical use, $Bi_4Ge_3O_{12}$ (BGO). BGO has been applied to radiation detection in which high stopping power for high-energy photons is necessary, because the density of BGO is high (7.13 g cm^{-3}). It should be noted that self-activated-type scintillation occurs in BGO [17], i.e., the luminescence occurs in the entire crystal of BGO, in contrast to the impurity-doped scintillators, in which the concentration of the luminescence centers is several mol% at most. Thus, at

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high LET, we expected a dependence of the decay and the rise on LET, because the closely located excited states at the luminescence sites can be achieved in the self-activated scintillator at high excitation density. In this study, we report the dependence of the rise and decay of the scintillation temporal profiles of BGO on LET.

2. Experimental

Scintillation temporal profiles at different LETs were obtained using pulsed ion beams from an AVF cyclotron at TIARA, QST, Japan. Here, a scintillation temporal profile is defined as scintillation intensity as a function of time after excitation (or irradiation). The samples were irradiated with pulsed beams of 20-MeV H^+, 50-MeV He^{2+}, and 220-MeV C^{5+}, which were obtained using two types of timing filters. The intensity of the pulse was less than 10^4 ions/pulse and the repetition rate was less than 100 pulses/s. The details of the pulsed beam generation system are described elsewhere [18].

A BGO crystal (1 cm × 1 cm × 1 mm) was irradiated in air. The scintillation from the sample was detected with a photomultiplier tube (PMT; Hamamatsu R7400U). The detection signal from the PMT was delivered to a digital storagescope (DSO; Tektronix DPO 7104) and the DSO was triggered with timing signals supplied from the accelerator. The scintillation temporal profiles were obtained as averaged signals over 1000 pulses. The time resolution of the measurement system was ~2 ns at half-width at half-maximum. The origin of the time axis was taken as the peak time of the scintillation temporal profile from a plastic scintillator, BC-400, in the same measurement setup. To obtain scintillation temporal profiles within different wavelength regions, optical filters were placed between the sample and the PMT. The change in the scintillation properties as a result of irradiation during the measurements was negligible. Further details of the measurement system are described in our previous paper [19]. The scintillation spectra were also obtained using an optical multichannel detector (Ocean Optics, USB-4000).

3. Results and discussion

Fig. 1 shows the LETs estimated with the SRIM code [20]. The LET is higher for heavier ions under the experimental conditions of this study and the LET ranges over two orders of magnitude.

Fig. 2 shows the scintillation spectrum under irradiation of 50-MeV He^{2+}. A broad band is observed at ~500 nm. This spectrum is consistent with those under photoexcitation [17] or X-ray irradiation [21]. The observed spectra were similar under irradiation of the other ions.

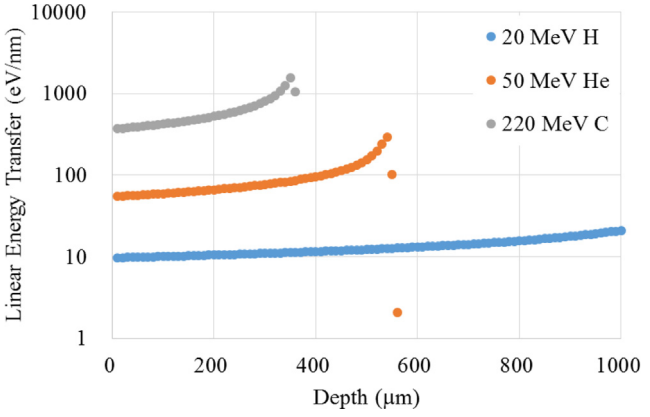

Fig. 1. Linear energy transfers estimated with the SRIM code.

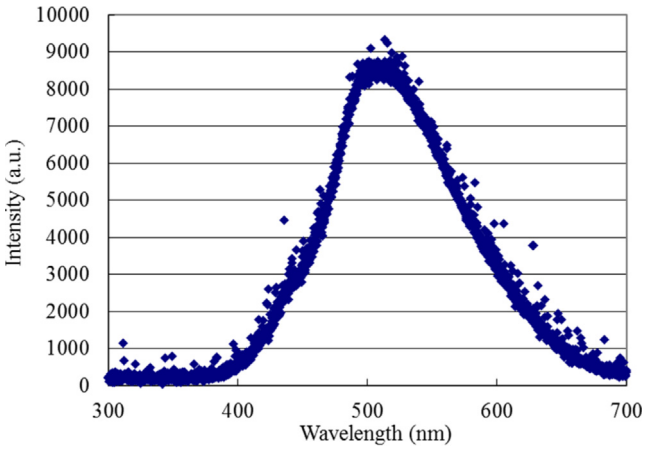

Fig. 2. Scintillation spectrum of BGO under irradiation of 50-MeV He^{2+}.

Fig. 3 shows the scintillation temporal profiles under irradiations of 20-MeV H^+, 50-MeV He^{2+}, and 220-MeV C^{5+} in the rise and decay regions. Firstly, it is evident that the rise is faster at higher LET. This same trend has been observed with a Li-glass scintillator [14], Ce-doped $LiCaAlF_6$ [15], and Ce-doped Gd_2SiO_5 [16]. Secondly, it can be observed that the decay s also faster at higher LET. In particular, for the irradiation of 220-MeV C^{5+}, a fast component appears, and the decay at longer than 200 ns is similar to that of the other ions. This is in contrast to the case of the Li-glass scintillator and Ce-doped $LiCaAlF_6$, for which no significant LET dependence was observed in the decay region. This can be attributed to

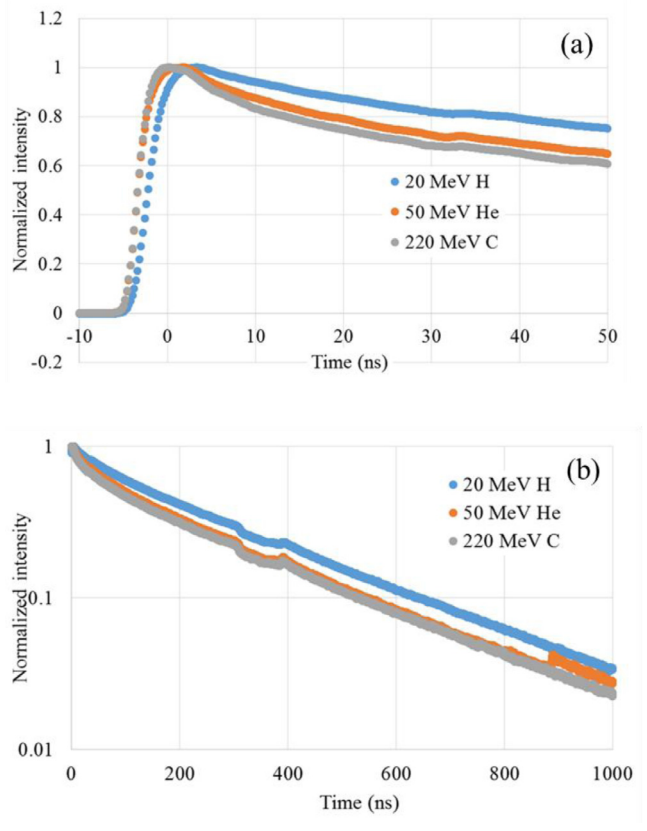

Fig. 3. Scintillation temporal profiles under irradiations of 20-MeV H^+, 50-MeV He^{2+}, and 220-MeV C^{5+} in (a) short and (b) long time range. The profiles are normalized to their peak intensities.

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