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Effects of Excitation Density on Scintillation Dynamics of $CdWO_4$

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The scintillation temporal profiles of $CdWO_4$ at different linear energy transfers (LETs) were analyzed in this study. The profiles were obtained using pulsed ion beams with different LETs in $CdWO_4$. In the temporal profiles, faster rise and decay were observed at higher LETs. The faster rise at higher LETs is attributed to the competition between the quenching due to excited-state interactions and the energy transfer from the host to the emission sites, i.e., the WO_6^{6-} complex. The faster decay is explained in terms of the competition between quenching due to excited-state interactions and radiative decay at the emission sites. These results, along with similar findings obtained for another self-activated scintillator, $Bi_4Ge_3O_{12}$, clearly indicate that self-activated scintillators exhibit scintillation temporal profiles that are significantly dependent on LET.

1. Introduction

High-energy ions deposit their energy densely in condensed matter, resulting in high-density electronic excitation. As a result, columnar defects along the trajectories of the high-energy ions are observed in insulators.(1) In addition to the permanent effects, the dense electronic excitation results in a response of radiation detectors; this response depends on the excitation density or the linear energy transfer (LET), which is defined as the deposited energy per unit length of the trajectory.

Among radiation detectors, phosphor-based detectors generally have an LET-dependent response. In the case of radiation measurements of high-energy ions, the LET dependence of the response of the phosphor should be accounted for in dosimetry. From this viewpoint, the LET-dependent response of the storage phosphors used as dosimeters has long been an issue in accurate dosimetry around the Bragg peak in particle therapy.(2–4) Many researchers have studied the LET-dependent response of storage phosphors based on various insulators doped with luminescence centers or centers for trapping electrons or holes.(5–7) Because storage

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phosphors store the energy deposited by ionizing radiation as metastably trapped electrons and holes at different sites, the LET dependence has been discussed in terms of the trapping process.

Another aspect of the LET-dependent response of phosphor-based radiation detectors is its application in pulse shape discrimination used for detecting gamma rays and neutrons in scintillation detectors. Neutrons are generally detected via nuclear reactions involving nuclides with large reaction cross sections, such as ^{3}He, ^{6}Li, ^{10}B, and several isotopes of Gd. Nuclear reactions of neutrons with ^{6}Li or ^{10}B, which are often used in phosphors for neutron detection, result in the emission of high-energy ions, such as tritons, alpha rays, and ^{7}Li, whose LETs are significantly higher than those of gamma rays. For scintillators with LET-dependent scintillation temporal profiles, gamma ray and neutron detection events have different scintillation temporal profiles, resulting in different detection signal shapes. Hence, the detection events of gamma rays and neutrons can be differentiated on the basis of the difference in the shape of the detection signal. Scintillators such as Li glass scintillators,[8–10] Ce-doped $LaCl_3$,[11] plastic scintillators,[12] and Ce-doped $LiCaAlF_6$[13] have been used for pulse shape discrimination. In general, scintillators found to have LET-dependent scintillation temporal profiles have been used for the aforementioned purpose. However, designing materials exhibiting such responses has been challenging because of the limited information available regarding the mechanism responsible for LET dependence.

To understand the mechanism of LET dependence, scintillation temporal profiles of various systems, e.g., some insulators including self-activated scintillators and doped scintillators, have been analyzed by several researchers. Kimura *et al.* developed a system for measuring scintillation temporal profiles with a time resolution of less than 100 ps using a time-correlated single-photon counting technique coupled with single-ion detection.[14] They analyzed the scintillation temporal profiles of many insulators under heavy-ion irradiation.[15–19] They also analyzed the scintillation temporal profiles of CdS[20] and Al_2O_3 in the vacuum ultraviolet region[21] in collaboration with our group. Other groups have measured the scintillation temporal profiles of organic scintillators, such as liquid scintillators,[22] naphthalene,[23] and polystyrene.[24]

Recently, our group has developed a system for measuring scintillation temporal profiles using pulsed ion beams generated from a cyclotron.[25] We have studied several Ce-doped scintillators, including $LiCaAlF_6$,[26] Li glass,[27] Gd_2SiO_5,[28] $Gd_3Al_2Ga_3O_{12}$,[29] and $Ca_3B_2O_6$.[30] In these cases, the quenching due to excited-state interactions competes with the energy transfer from the host to Ce^{3+} ions,[26–28] or multiple excited states compete for energy transfer to a nearby Ce^{3+} ion.[26,29,30] LET-dependent scintillation temporal profiles have also been observed in low-dimensional semiconductor scintillators based on organic–inorganic layered perovskite-type compounds, in which the interaction of the Wannier excitons in the inorganic layers plays an important role in the LET dependence.[31]

In this study, we focus on self-activated scintillators. In our previous study, we reported that both the rise and decay of the scintillation temporal profile of $Bi_4Ge_3O_{12}$ depend significantly on LET.[32] The LET-dependent rise and decay can be attributed to the competition between quenching due to excited-state interactions and energy transfer to emission sites, and to the radiative transition at the emission sites, respectively. To confirm this hypothesis, we studied

another self-activated scintillator, $CdWO_4$, following a methodology similar to that used in a previous study.[32]

2. Materials and Methods

The sample used for this study was a $CdWO_4$ single crystal with dimensions of $10 \times 10 \times 1$ mm^3. The scintillation temporal profiles were obtained using our original setup at TIARA, QST, Japan.[25] The sample was irradiated in air with pulsed beams of 20 MeV H^+, 50 MeV He^{2+}, and 130 MeV C^{5+}, which were obtained from an azimuthally varying field (AVF) cyclotron using a combination of choppers.[33] The scintillation photons from the sample were detected using a photomultiplier tube (PMT; Hamamatsu, R7400U). The output signals of the PMT were delivered to the measurement room outside the irradiation room and were recorded using a digital oscilloscope (Tektronix, DPO 7104), which was triggered by the timing signals supplied from the accelerator. The detection signals over 1000 pulses were averaged to obtain the scintillation temporal profiles. The overall time resolution of the measurement system was ~2 ns at half width at half maximum.[25] The time origin ($t = 0$) was set as the peak timing of the scintillation temporal profiles of a plastic scintillator, BC-400, for different ions under the same conditions as those of the measurements of the sample. The scintillation spectra of the samples were recorded using a multichannel spectrometer (Ocean Optics, USB-4000). The LETs of the ions in the sample were estimated using the SRIM code.[34]

3. Results and Discussion

The LETs of the different ions used for the irradiation of the sample estimated using the SRIM code are shown in Fig. 1. The LET was found to be higher for heavier ions, and it changed by over two orders of magnitude among the ions.

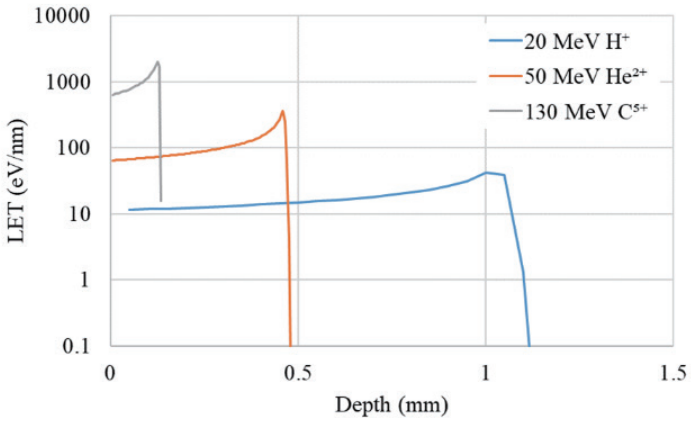

Fig. 1. (Color online) LETs estimated using the SRIM code.

The scintillation spectrum of the $CdWO_4$ sample under the 50 MeV He^{2+} irradiation is shown in Fig. 2. An emission peak was observed at 490 nm, which is consistent with a previously reported emission spectrum and was attributed to the emission of the WO_6^{6-} complex.[35]

The scintillation temporal profiles of the $CdWO_4$ sample irradiated with 20 MeV H^+, 50 MeV He^{2+}, and 130 MeV C^{5+} in a short time range of up to 50 ns are presented in Fig. 3. The temporal profiles were normalized to their peak intensities. The rise in the temporal profile for 130 MeV C^{5+} is much faster than those observed for the other ions. According to this result, excited-state interactions occur for 130 MeV C^{5+} after excitation by the ions. In contrast, the influence of the excited-state interactions on the scintillation temporal profiles is much less significant at lower LETs.

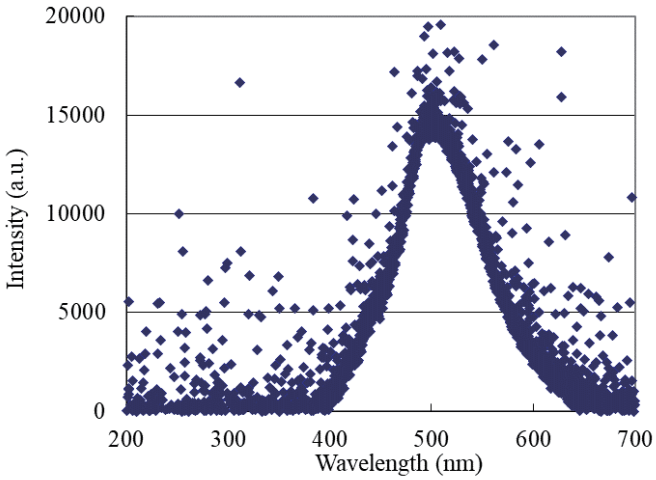

Fig. 2. (Color online) Scintillation spectrum of the $CdWO_4$ sample under 50 MeV He^{2+} irradiation.

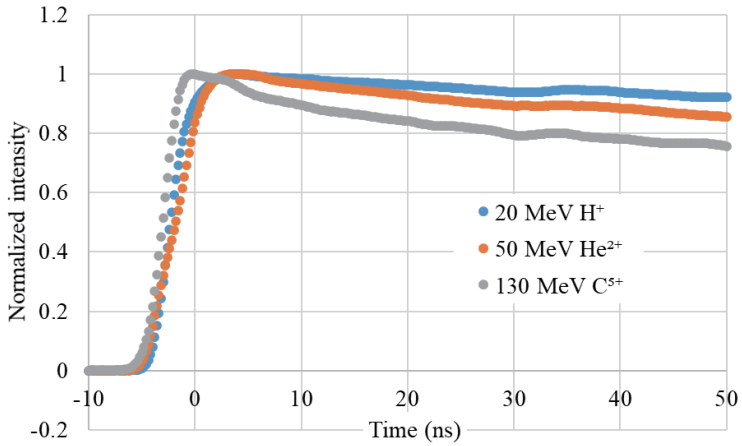

Fig. 3. (Color online) Scintillation temporal profiles of the $CdWO_4$ sample for 20 MeV H^+, 50 MeV He^{2+}, and 130 MeV C^{5+} irradiation in a short time range of up to 50 ns. The profiles are normalized to their peak intensities.

Figure 4 shows the scintillation temporal profiles of the $CdWO_4$ sample irradiated with 20 MeV H^+, 50 MeV He^{2+}, and 130 MeV C^{5+} in an intermediate time range of up to 300 ns, which were recorded to observe the initial decay behavior. A small reflection noise was observed at ~30 ns. The initial decay up to ~50 ns was faster for a higher LET. Contrary to the case of the rise, the initial decay gradually became faster with increasing LET.

The scintillation temporal profiles of the $CdWO_4$ sample for the 20 MeV H^+, 50 MeV He^{2+}, and 130 MeV C^{5+} irradiation in a long time range of up to 1500 ns are presented in Fig. 5. The vertical axis is set to a logarithmic scale. The trends in the decay of the scintillation temporal profiles are not straight lines, which suggests that the decay does not follow first-order kinetics. The decay was faster for the higher LET throughout the 1500 ns. This indicates that the excited-state interactions occur even at a long elapsed time of up to 1500 ns. The overall decay results are consistent with the scintillation decay behaviors observed for gamma rays and alpha rays.[36]

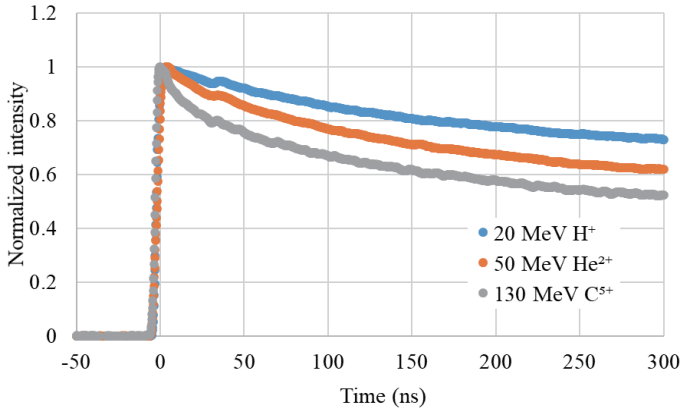

Fig. 4. (Color online) Scintillation temporal profiles of the $CdWO_4$ sample for 20 MeV H^+, 50 MeV He^{2+}, and 130 MeV C^{5+} irradiation in an intermediate time range of up to 300 ns. The profiles are normalized to their peak intensities.

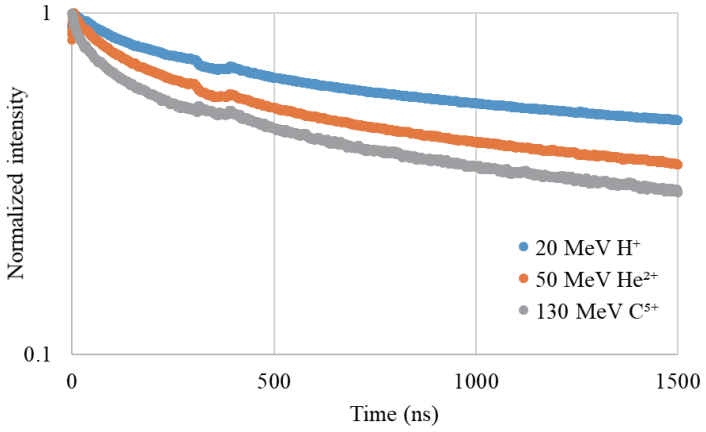

Fig. 5. (Color online) Scintillation temporal profiles of the $CdWO_4$ sample for 20 MeV H^+, 50 MeV He^{2+}, and 130 MeV C^{5+} irradiation in a long time range of up to 1500 ns.

The LET dependence of the rise and decay of the scintillation temporal profiles is discussed herein. Because the emission is attributed to the emission of the WO_6^{6-} complex,[35] the rise corresponds to the formation of the excited WO_6^{6-} complex. The rise for ~5 ns in the scintillation temporal profiles observed for 20 MeV H^+ and 50 MeV He^{2+}, i.e., at low LETs, strongly indicates that one electron–hole pair or the excited state of the host transfers its energy to the WO_6^{6-} complex in ~5 ns. As the α/β ratio of $CdWO_4$ is less than 0.25,[36] the faster rise in the scintillation temporal profiles at a higher LET may be explained on the basis of quenching. A similar behavior of the rise in the scintillation temporal profile has been observed for another self-activated scintillator, $Bi_4Ge_3O_{12}$,[32] and for some doped scintillators, such as Ce-doped $LiCaAlF_6$,[27] Ce-doped Li glass,[28] and Ce-doped Gd_2SiO_5.[29] Such behavior can be explained in terms of the competition between quenching due to excited-state interactions and energy transfer from the host to the emission sites.

The faster decay of the scintillation temporal profile at the higher LET can also be explained in terms of the competition between the quenching due to the excited-state interaction and the radiative transition at the emission sites. Such behavior has been observed under intense laser irradiation in similar compounds, such as $PbWO_4$ [37] and $ZnWO_4$.[38] This observation is in clear contrast to the behavior observed for Ce-doped $LiCaAlF_6$,[26] Ce-doped Li glass,[27] and Ce-doped Gd_2SiO_5,[28] where the decay of the 5d-4f emission of Ce^{3+} ions is almost independent of LET. The difference in the two behaviors observed can be explained as follows. In Ce-doped samples, the decay behavior reflects the decrease in the number of excited states of Ce^{3+} ions. The concentration of emission sites, i.e., Ce^{3+} ions, is low, and the distance between the Ce^{3+} ions in their excited states is high. Hence, the interactions between the excited Ce^{3+} ions are negligible. In contrast, in the self-activated scintillators, such as $Bi_4Ge_3O_{12}$ and $CdWO_4$, the concentration of emission sites (Bi^{3+} ions or the WO_6^{6-} complex) is high. Therefore, the excited states are located close to each other, resulting in interaction between the excited states at a high excitation density or LET.

The effects of LET on the scintillation temporal profiles are more significant in self-activated scintillators than in doped scintillators. From the viewpoint of pulse shape discrimination of detection events of gamma rays and neutrons, self-activated scintillators are advantageous because the rise and decay of their scintillation temporal profiles are significantly dependent on LET, which facilitates pulse shape discrimination.

4. Conclusions

We investigated the effects of LET on the scintillation temporal profiles of $CdWO_4$ single crystals. We obtained scintillation temporal profiles at different LETs under the irradiation of 20 MeV H^+, 50 MeV He^{2+}, and 130 MeV C^{5+}. On the basis of Monte Carlo simulation using the SRIM code, the LET was estimated to be higher for the heavier ions in this study. The rise and decay of the scintillation temporal profiles were faster at a higher LET. The faster rise at a higher LET is consistent with the results of some doped scintillators studied earlier and of another self-activated scintillator, $Bi_4Ge_3O_{12}$. This behavior is explained in terms of the competition between quenching due to excited-state interactions and energy transfer from the host to the emission

sites. The faster decay at a higher LET is explained by the competition between quenching due to excited-state interactions and radiative transition at the emission sites. These results, along with observations for $Bi_4Ge_3O_{12}$, indicate that the scintillation temporal profiles of self-activated scintillators are more LET-dependent than those of doped scintillators.

Acknowledgments

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