SCINTILLATION DEGRADATION OF YAG: CE UNDER LOW-ENERGY ION BOMBARDMENT

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Abstract

We report on the degradation of the scintillation yield of single crystal YAG: Ce under He^+ irradiation at low energy. The decrease of the luminescence intensity as a function of fluence can be analyzed by the Birks model. The results indicate that the scintillator degrades fast and exhibits poor scintillation efficiency for low-energy irradiation. A possible degradation mechanism is proposed.

INTRODUCTION

YAG: Ce scintillators have been widely employed as accelerator beam diagnostics or positioning system due to their high output, a decay time of less than 100 ns and long lifetime in a high radiation environment [1]. The scintillation spectrum of YAG: Ce is constituted by a broad peak at about 550 nm coming from the recombination of excitons near a Ce^{3+} ion. The scheme of the scintillation is presented in Fig. 1. A cerium ion has an electron configuration of $[Xe]4f^1$. The 4f electron can be excited into the empty 5f shell. When a Ce^{3+} is doped in a lattice of YAG, the crystal field splits the 4f into two and the 5d level into five sublevels. Once incident particles and the subsequent δ-rays ionize the atoms in the crystal, electrons can be excited into the conduction band and leave behind an equal number of holes in the valence band. As these electron-hole pairs eventually thermalize and drift through the lattice, they preferentially recombine at the local activator sites as a result of the 5d-4f electric dipole transition. The photons are emitted usually due to the transitions from the lowest 5d level to the two 4f levels [2].

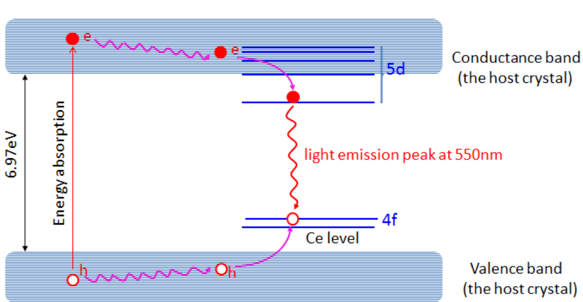

Figure 1: Schematics of the scintillation process in YAG: Ce. The electron-hole pair generated by the interaction of incident particles is being transported and trapped at the Ce^{3+} site. The trapped exciton recombines to emit visible light through the 5d-4f transition.

However, it has been known that the induced radiation damage in scintillators leads to the degradation of the luminescence intensity. From our observation, this degradation is particularly pronounced under low-energy ion bombardment, which is usually applied in beam monitoring systems of a low-energy accelerator. Therefore, the investigation of the decay mechanism is of importance in order to obtain better resolution of beam profile monitors for heavy ion beam injectors. In this paper, we report on the scintillation response and timing behavior of the YAG: Ce single crystal under He^+ irradiation at low energy. The degradation caused by incident ion fluence has been analyzed in terms of half brightness dose [3].

EXPERIMENTAL

The irradiation of the YAG:Ce scintillator and the simultaneous measurement of its scintillation yield have been performed at the rare isotope ReAccelerator (ReA) facility at the National Superconducting Cyclotron Laboratory (NSCL) [4] at Michigan State University (MSU). He^+ beams with energies between 28 and 58 keV (varied in 10 keV steps) were used for these tests. In order to suppress the thermal effect of the irradiated scintillator, the beam current measured by a Faraday cup was limited to intensities of less than 500 pA. The beam spot for each test on the scintillator was steered to a different location on the scintillator to make sure the beam impinged upon a fresh surface. A fine mesh was attached to the scintillator surface to prevent charge build-up caused by the ion beams. The angle between the scintillator and the beam axis was chosen at 45° for the best light collection as shown in Fig. 2. The scintillation light as a function of the irradiation time was recorded outside of the vacuum chamber by a CCD camera equipped with 752×480 resolution. The camera was located perpendicular with respect to the beam axis.

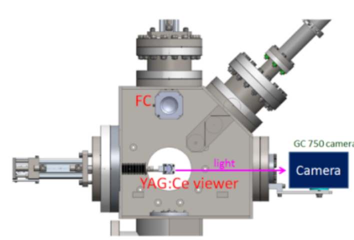

Figure 2: The setup of the YAG: Ce scintillator at the rare isotope ReAccelerator. After the beam was tuned into the Faraday cup and a beam current measurement was acquired, the Faraday cup was pulled up and the scintillator plate was inserted into the beam. The emitted light was observed with a camera.

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RESULTS AND DISCUSSION

An example for the degradation of the luminescence intensity under continuous He^{+} bombardment can be seen in Fig. 3. After a total irradiation of 192 s, the scintillation becomes much weaker due to some damaging effect. Two mechanisms may play a dominant role in the degradation of the luminescence due to ion beam bombardment. The luminescence center Ce^{3+} might be changed to Ce^{4+} by energy transfer [5]. The emission band of Ce^{3+} is overlapped by the absorption band of Ce^{4+} which decreases light output. Another mechanism is the most common damage caused by the knock-out of oxygen atoms. The ion bombardment can create the electron-hole pairs and the oxygen vacancies in YAG: Ce scintillators. The oxygen vacancies have three kinds of charge states: the one-electron F^{+} center, the two-electron F^{0} center and the three-electron F^{-} center. The F^{-} center can absorb the photons mainly emitted from the Ce^{3+} sites and its concentration is enhanced by ion irradiation. The absorption band caused by the formation of the F^{-} center reduces the crystal's light attenuation, and hence the light output [6].

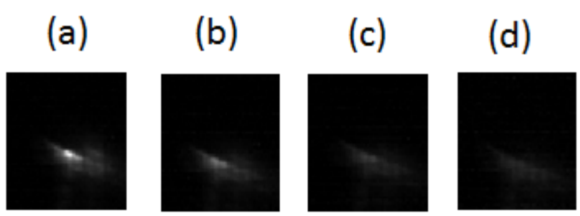

Figure 3: The degradation of YAG: Ce after a total irradiation of (a) 0 s, (b) 25 s, (c) 101 s and (d) 192 s at incident ion energy E=58 keV and beam current I=394 pA.

The degradation of the scintillation yield for YAG: Ce under He^{+} irradiation can be analyzed by the Birks model [3]. In 1951, Birks and Black published an experimental formula to describe the luminescence efficiency of the scintillator bombarded by Helium ion beams. This empirical formula has been tested for many scintillator materials [7]. The Birks model was derived from the idea that the degradation of the luminescence is due to the absorption of photons by damaged molecules. For an analyzed area of the scintillator, the incident particle fluence as a function of the inversely normalized scintillation yield can be described by

$$N = \frac{1}{\sigma_d} \cdot \ln\left(1 + \frac{\frac{I_0}{I} - 1}{k}\right) \tag{1}$$

where I and I_0 represent the scintillation intensity for a given fluence and the initial intensity, respectively. The particle fluence N (mm^{-2}) is the total number of beam particles per mm^{-2} striking the crystal at a given time expressed as [8]

$$N = \frac{\Delta Q}{qeA} \tag{2}$$

where ΔQ is the total integrated charge accumulated on an analyzed area, q is the charge state, and A is the area of the analyzed pixels with the maximum of initial brightness. The errors in determining the fluence can be contributed to the calculation of the total initial brightness on the beam spot and the measurement of time and current.

There are two fitting parameters in the Birks model: k and σ_d. The relative exciton capture probability k is the ratio of the number of excitons captured by a damaged molecule to an undamaged molecule. If q_0 is the number of scintillator molecules in the irradiation volume per mm^2 and each beam particle damages p molecules, the damage cross section σ_d is defined as $^{p}/_{q_0}$. Each set of experimental data at a specific beam energy and current has been fitted with the Birks model by determining the least chi-square χ^2 in order to evaluate k and σ_d. As an example of the experimental data and the fit of the particle fluence versus the inversely normalized scintillation intensity ($^{I_0}/_{I}$) under 48 keV He^{+} irradiation is shown in Fig. 4. The variations of k and σ_d in dependence of the beam energy are presented in Fig. 5.

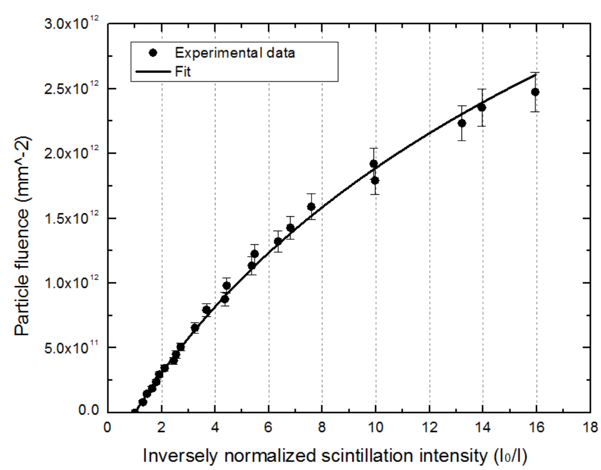

Figure 4: The particle fluence vs. the inversely normalized scintillation intensity at incident He^{+} energy E=48 keV and beam current I=200 pA. The dots with error bars are the experimental points and the solid line shows the fit of the Birks model.

As it can be seen in Fig. 5a, we do not find a significant energy dependence of k within the experimental uncertainty. In addition, the nearly constant value of k is considerably small compared to other scintillators [9]. It reveals that the YAG: Ce scintillator exhibits much higher resistance to radiation damage. On the other hand, the damage cross section slightly decreases with the ion energy (figure 5b). From our analysis for the oxygen vacancies created per incident ion and the beam penetration depth into the scintillator using the SRIM

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code [10], it appears that although higher incident ion energy can create more defect centers, the damage cross section is reduced because the deeper penetration activates more luminescence centers.

The degradation of the scintillation intensity can be evaluated in terms of the half brightness dose $N_{1/2}$ defined as the amount of the particle fluence required to degrade the luminescence to one half of its original value. The value of $N_{1/2}$ deduced from our measurements is presented in Fig. 6, showing that reducing the ion energy lowers the half brightness dose. The lower the incident energy the faster the YAG: Ce scintillator degradation of its luminescence.

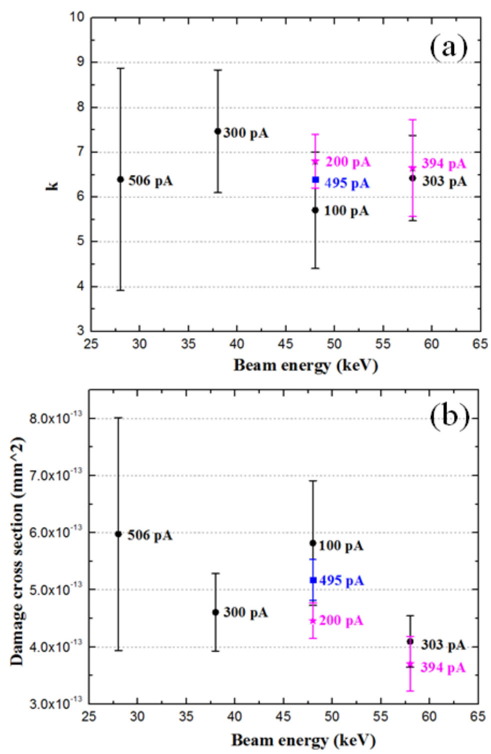

Figure 5: The relative exciton capture probability and the damage cross section as a function of the He^+ energy. The error bars are the experimental uncertainty. Three symbols are used to represent different tests with different current at the same energy.

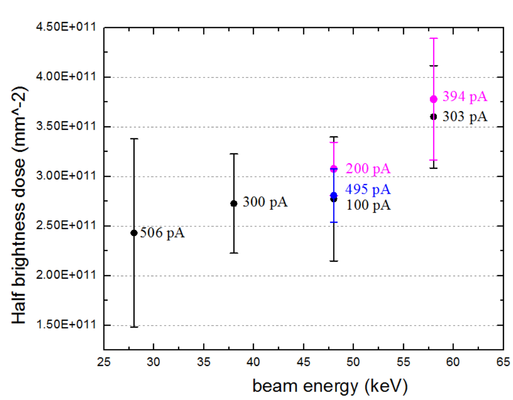

Figure 6: The half brightness dose vs. the He^+ energy.

CONCLUSIONS

We have systematically measured the degradation of the scintillation yield for YAG: Ce under He^+ irradiation. Using the Birks model, the damage cross section and the half brightness dose was estimated for single charged helium ions in the energy range of 28keV to 48keV. The higher the energy of the incident ions the deeper they can penetrate into the scintillator volume and activate more luminescence centers to emit photons. As a result, the scintillation degrades less and the half brightness dose increases with ion energy.

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