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An ideal scintillator – ZnO:Sc for sub-nanosecond pulsed radiation detection

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ABSTRACT

ZnO-based scintillators are particularly well suited for use as ultrafast pulsed radiation detectors which have shown broad application prospects in various fields such as the inertial confinement fusion (ICF) diagnosis, the nuclear reaction mechanism, etc. Using the hydro-thermal method, a ZnO single-crystal doped with Scandium (ZnO:Sc) sample was prepared. As a new ZnO-based scintillator, the scintillation characteristics of ZnO:Sc have not been reported previously. In this paper, optical and scintillation characteristics of ZnO:Sc single-crystal were studied. Also a scintillation detector based on ZnO:Sc was designed. Excited by the alpha-particle, the rise time of ZnO:Sc detectors was from 162.5 to 170.7 ps, and the fall time was from 300.4 to 328.8 ps.

1. Introduction

As a direct wide band-gap semiconductor (3.3 eV at room temperature), zinc oxide (ZnO) has demonstrated room temperature sub-nanosecond photo- and radio-luminescence when doped with n-type dopants such as indium, gallium, or Iron. Due to outstanding timing resolution, medium density (5.6 g/cm^3), high radiation resistance, and the exclusion of organic materials, ZnO-based scintillators are particularly well suited for use as ultrafast pulsed radiation detectors (for neutron, gamma-ray, X-ray, alpha-particle, etc.) which have potential applications in various fields such as the inertial confinement fusion (ICF) diagnosis, the International Thermonuclear Experiment Reactor (ITER) diagnosis [1], astrophysics [2], radiation imaging [3], the nuclear reaction mechanism, homeland security, etc.

The optical and scintillation characteristics of ZnO change with different dopants and dopant concentrations. Since 1960s [4,5], ZnO-based materials have been known as ultrafast scintillators; lots of research has been done on ZnO doped with Gallium or Indium mostly, as well as Fe, Mn, Co, Ni, V, etc. [6–9]. Unfortunately, the early ZnO-based study samples are always powders or films, which are difficult to assemble for radiation detectors and because of self-absorption, the light yield from powders or films is often less than satisfactory. Those drawbacks have restricted the further studies of scintillation characteristics of ZnO-based scintillators in the past decades, until ZnO-based single-crystal of sufficient size has become available only recently [10]. ZnO-base single-crystals, such as ZnO:Li, ZnO:Mg, ZnO:Ga, etc., were tested as a kind of alpha-particle detector [10–14]. ZnO:In crystal with 3-ps response time was tested for survey of X-ray free electron laser pulse [15].

The ZnO single-crystal doped with Scandium (ZnO:Sc) is a new member of the ZnO family, and its scintillation characteristics have not been reported before. Compared with other doped ZnO, the size of the ZnO:Sc single-crystal can be grown larger relatively, which is obviously a huge advantage. To better adapt the ZnO-based scintillators to the potential application fields and to improve the radiation detection capabilities of ZnO-based detectors, the optical properties and scintillation characteristics of ZnO:Sc were studied in this paper. Furthermore a ZnO:Sc scintillation detector was designed and tested.

2. Optical properties

The ZnO:Sc single-crystal was grown by China Nonferrous Metal Guilin Research Institute of Geology for Mineral Resource, using the hydrothermal method [16]. Fig. 1 shows the photograph of a ZnO:Sc single-crystal tested sample. The crystal is transparent and faint yellow, with a dimension of 15 mm × 15 mm × 0.3 mm.

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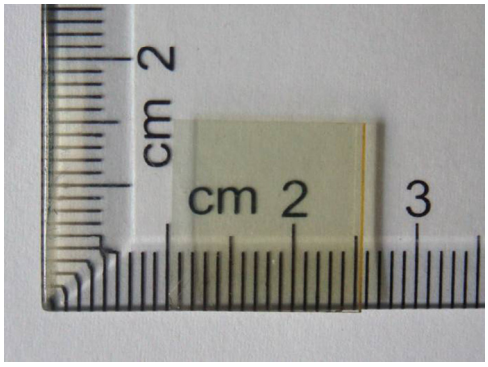

Fig. 1. ZnO:Sc single-crystal grown by the hydrothermal method. The dimension is 15 mm × 15 mm × 0.3 mm.

Table 1
Impurity analysis of ZnO:Sc crystal determined by ICP-MS.

Unit: ppm								
Impurities	Al	Fe	K	Li	Cu	Si	Sc	Au
+c Sector	0.82	10.7	0.21	1.1	7.4	< 0.3	8.3	0.85
−c Sector	0.94	13.8	0.40	2.1	7.5	< 0.3	11.9	0.17

The impurity concentrations were analyzed by the inductively coupled plasma mass spectrometry (ICP-MS). The results are shown in Table 1, the concentration of Sc^{3+} was just several ppm in both +c and −c sectors.

To compare the characteristics of ZnO:Sc with other ZnO-based scintillators, a commercial ZnO:Ga single-crystal product was also tested as an reference sample. The ZnO:Ga sample of 10 mm × 10 mm × 0.5 mm dimension was grown by MTI Corporation (CA, US) [17], using a melt-growth process.

The room-temperature photoluminescence spectra of ZnO:Sc and ZnO:Ga samples were made using a He–Cd laser as the exciting source at the wavelength of 325 nm (Fig. 2). The two emission peaks of ZnO:Sc sample were approximately centered at 377 nm and 556 nm. The spectrum of ZnO:Ga showed only one emission peak approximately centered at 377 nm and no visible emission was observed.

For the ZnO:Sc sample, the former ultraviolet emission is related to the exciton-based emission and the latter emission is usually ascribed to an ionized oxygen vacancy and an oxygen interstitial [18]. For undoped ZnO crystal, the ultraviolet peak was reported at 377 nm [19]. The undoped ZnO, ZnO:Sc and ZnO:Ga are unanimous in the ultraviolet peak wavelength. Neal [13], and Kano [20], reported that the ultraviolet peak wavelength shifts towards longer wavelengths when ZnO crystal is doped with dopants. However this phenomenon had not been observed in our study.

The optical transmission spectra of ZnO samples were measured using a UV-3101 ultraviolet–visible (UV) scanning spectrometer (SHIMADZU, Japan). All measurements were made at room temperature with a spectrum scanning range of 160–800 nm. Fig. 3 shows the transmittance spectra of ZnO:Sc and ZnO:Ga. The absorption edge of both ZnO samples appears at 387 nm, but the shape of transmittance spectra curve of ZnO:Sc and ZnO:Ga is different in spectral range of 387–550 nm. While the transmittance values of ZnO:Ga are larger in that wavelength range, the transmittance of ZnO:Ga increased much rapidly than that of ZnO:Sc at first as the incident light wavelength increases, and then the increase trend slows down. Above 450 nm, both samples show excellent transmittancy and the transmittance is up to 70–80%. Above 550 nm, the transmittance became almost constant at about 80%.

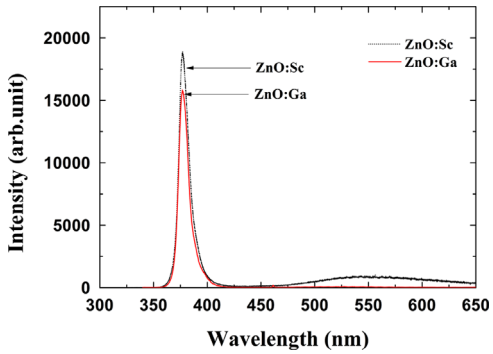

Fig. 2. Photoluminescence spectra of ZnO:Sc and ZnO:Ga single-crystals.

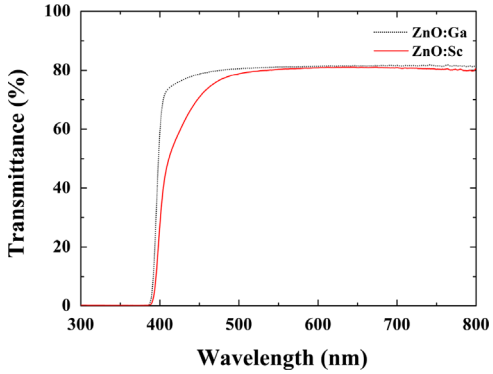

Fig. 3. Transmittance of ZnO:Sc and ZnO:Ga single-crystals.

3. Alpha-particle excited response

3.1. Alpha excitation

The radioluminescence was measured under the 9.9×10^3 Bq ^{241}Am radioisotope alpha source, the activity region of which was a circle with a diameter of 7 mm. The normalized energy of the alpha-particle was about 5.50 MeV. The alpha source clung to ZnO:Sc sample and the alpha-particle passed through ~1 mm of air for measurement. The incident alpha-particle range in the ZnO:Sc sample was calculated with the code SRIM (Stopping and Range of Ions in Matter) 2003. According to the calculation, the normalized alpha-particle range in ZnO:Sc was about 15.4 μm which was much smaller than the thickness of ZnO:Sc sample. This means that the incident alpha-particle can be fully absorbed by the sample and the thickness of the sample was enough to detect alpha-particles in the following experiment.

3.2. Alpha-particle excited spectrum

The schematic diagram of the experimental setup used to measure the ZnO:Sc single-crystal energy spectrum excited by alpha-particle is shown in Fig. 4. The single-crystal ZnO:Sc sample was coupled to the light entrance window of PMT 9815B [21] of which photocathode is made of Bi-Alkali. The quantum efficiency at peak (~360 nm) of PMT 9815B is about 30% and its spectral range is 290–630 nm. The high voltage was supplied by Stanford PS350 with −1700 V, and the signals were read out from the anode of the PMT. Then, the signals passed a preamplifier ORTEC 9305 and a shaping amplifier ORTEC 570 with 1 μs shaping time. After being converted to digital signals by a multi-channel analyzer EASY-MCA-8K using 8192 channels data acquisitions, they were recorded on a computer. The pulsed time response was also observed coarsely by an oscilloscope, Tektronix DPO 4104; the accurate data will be measured in the next experiment with the