Properties study of ZnO:Ga crystal on pulsed radiation detections*

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Abstract In this paper, properties on pulsed radiation detections of ZnO:Ga crystal grew by a magnetron sputtering method were studied. The time response to pulsed laser，pulsed hard X rays and single α particles, the energy response to pulsed hard X ray, the scintillation efficiency to γ rays, the response to pulsed proton, and the relations of the light intensity varied with the proton energy were measured and analyzed in detail. Results show that the ZnO:Ga crystal has potential applications in the regime of pulse radiation detection .

Key words ZnO:Ga, inorganic scintillator, radiation detection, time response, energy response, luminescence efficiency

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1 Introduction

Since 1968 when Luckey [1] reported on the excellent performance of gallium-doped zinc oxide (ZnO:Ga) prepared from a multicrystalline powder, researchers have demonstrated an interest in the optimization of the performance of the ZnO:Ga crystal. ZnO is a Ⅱ–Ⅳ compound semiconductor with a band-gap energy of 3.37 eV at room temperature, sub-nanosecond luminescence when doped with n-type dopants such as gallium and indium [2]. The ZnO:Ga scintillation has been developed for use with sealed tube DT neutron generators [3]. According to the Lawrence Berkeley Laboratory reports [4, 5], the density of the ZnO:Ga crystal is 5.61 g/cm^3, not deliquescent. When excited by X ray, the emission wavelength is 385 nm. The high luminous efficiency is 15000 photos/MeV, it is 0.4 times as large as that of the NaI:Tl crystal. The exponential decay time and the percentage of the ZnO:Ga crystal are 0.6 ns and 97.5%.

Thus it can be seen that the ZnO:Ga crystal is a new type of inorganic scintillator. The higher light output and the very fast time response are obvious characteristics. In addition, there is no hydrogen in the ZnO:Ga crystal, and the section that reacts to neutron is little. So it can be applied in ultra-fast pulsed gamma detection, fusion neutron energy spectrum detection, fusion neutron time spectrum detection and plasma hard X ray diagnosis. In this paper, we have described the irradiation effect of the ZnO:Ga crystal to femtosecond laser, X ray, α particle, γ ray and proton, attained the time response and the energy response to different radiation particles, and analyzed the results and phenomena.

The ZnO:Ga crystal was prepared by a magnetron sputtering method using ZnO and 0.01% Ga_2O_3 as the starting materials in Xi'an Jiaotong University. The diameter was 50 mm and the thickness was 50 μm. The Al_2O_3 (0001) surface crystal served as substrate materials. Lastly, the samples were subjected to an oxygen anneal, and the temperature was about 800 ℃. Fig. 1 illustrates the transmittance of the ZnO:Ga crystal, the average is about 80% (without deducting the sapphire substrate absorption). Fig. 2 shows the fluorescence spectrum of ZnO:Ga crystal using 248 nm laser excitation.

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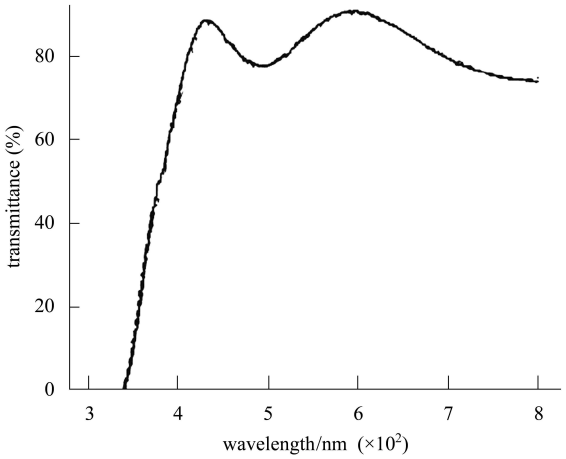

Fig. 1. The transmittance of the ZnO:Ga crystal.

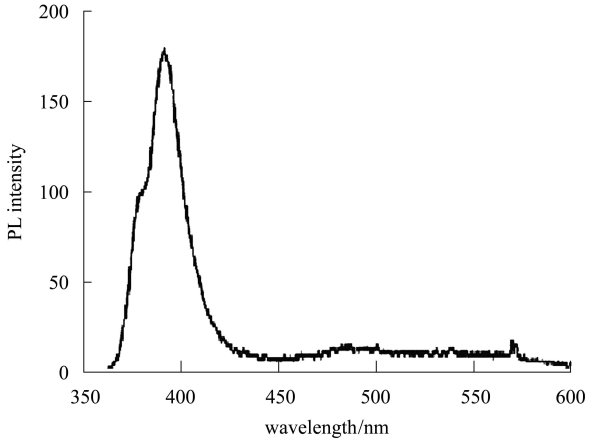

Fig. 2. The fluorescence spectrum of ZnO:Ga crystal.

It should be noted that the transmittance of the ZnO:Ga crystal that was annealed is slightly higher than that which was not annealed. The spectrum range is between 380 nm to 410 nm, and the scintillation light peak is at 390 nm.

2 Time performance

2.1 Time response to femtosecond laser

An ultrafast ultraviolet pulsed laser with a wavelength of 248 nm and pulse width of 500 ps was used as the light source in the measurement [6]. The time response of ZnO:Ga crystal to the pulsed laser was near Gaussian-type, which can be calculated from the root-mean-square value. The time response of the measuring system was composed of the response of the photoelectric tube, the signal cable, and the oscilloscope. The experimental results showed that the rise time of the system was 104.9 ps and the FWHM was 153.5 ps, and that of the ZnO:Ga crystal was 56.0 ps and 72.8 ps correspondingly if the system response was deduced. The respective scintillation peak wavelength of the ZnO:Ga crystal and ST401 scintillation are 390 nm and 423 nm, which are shown in Fig. 3 and Fig. 4 respectively.

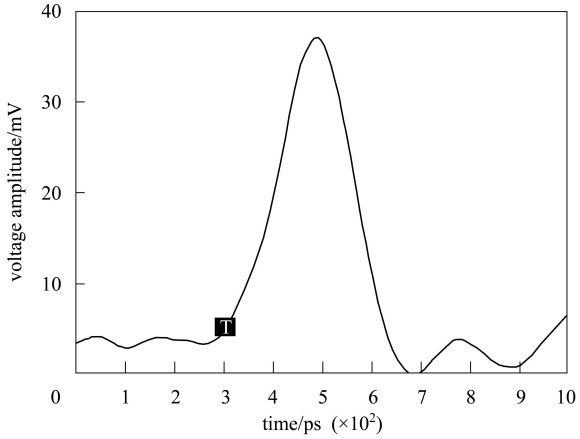

Fig. 3. The time response of the ZnO:Ga crystal to laser.

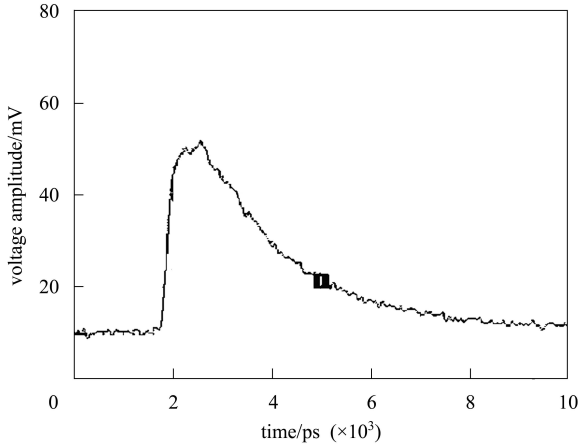

Fig. 4. The time response of the ST401 crystal to laser.

It should be noted that the better the quality of the crystal, the faster the time response. It can be seen that in Fig. 4 an obvious platform is generated by the slow descent of the trailing edge of the time response of the ST401 scintillation. However, the trailing edge of the time response of the ZnO:Ga crystal descends sharply as shown in Fig. 3, which indicates that the decay time constant is much smaller. In Fig. 3, the decay time between 90% of the peak value to 10% is 0.097 ns, so the decay time constant of the ZnO:Ga crystal is 0.044 ns, which is much smaller than the 0.7 ns that was reported in Ref. [4].

2.2 Time response to single α particle

The time response of the detector can be measured with a single charged particle due to its short track interacting with the ZnO:Ga crystal, while each

track is independent. As the flight of α particle is as short as a few centimeters in the air, the probe container must maintain low vacuum. ^{241}Am with intensity 12 Bq was used as the radiation source, and the energy of the α particle was 5.4 MeV. Time response of the ZnO:Ga crystal to single α particle is shown in Fig. 5.

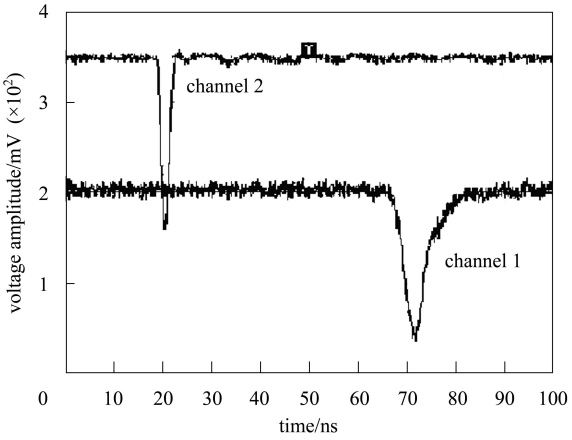

Fig. 5. The time response of the ZnO:Ga crystal to single α particle. The signal of channel 1 is the trigger pulse of EMI 9813 photomultiplier, and the signal of channel 2 is the waveform of an α particle detected by a microchannel photomultiplier (MCP).

The photons excited by the α particle in the ZnO:Ga crystal entered into the two photomultipliers simultaneously coincidently event. The two pulses should display on the oscilloscope at the same time with the time interval 51 ns, which is caused by the discrepancy of the transit time of the two photomultiplier. The peak voltage of the MCP varied significantly, dived to the hexagonal prism island structure of ZnO:Ga crystal. As a single α particle with different angles incidents on different crystal faces, the excited photon exits from different crystal faces with different departure angles, thus, the photon arrived at the photomultiplier with great discrepancy. This phenomenon has been validated in the detection of the fluorescence spectrum by optical fiber spectrometer. By deducting the time response of the oscilloscope, the photomultiplier, and the signal cable, the time response to the single alpha particle was gained. Results showed that the rise time was 324 ps and the FWHM was 686 ps.

2.3 Time response to pulsed hard X ray

By using the coincidence method, the time response of ZnO:Ga crystal to a fast rep-rate pulse hard X ray of the Northwest Institute of Nuclear Technology was measured. The rise time was 316 ps and the FWHM was 440 ps, which were not as good as those to laser, but much better than other ordinary crystals. The difference comes from the photo-translating system. In the laser experiment a photoelectric tube with a 50 ps response time was used, whereas in the X ray experiment, the response time of the photoelectric tube used was 350 ps, which caused much uncertainty in the measurement. In addition, the experiment on X ray is only a relative measurement, but that on laser is absolute.

The process of charge collection of heavy charged particles is different from the photon's equalizing ionization in the whole sensitive region, generally the energy is higher, and the plasma effect emerges. In theory, the plasma time of α particle is longer than that of the proton, while the proton's is longer than that of photon. Thus, the time response of α particle is better than proton, whereas the response of the proton is better than the photon, the rise time (t_r) and the peak half width (t_{FWHM}) of ZnO:Ga crystal are:

$$56.01\ \text{ps} \leqslant t_{FWHM} \leqslant 686.94\ \text{ps},\ 72.87\ \text{ps} \leqslant t_r \leqslant 324.88\ \text{ps}$$

3 Energy response to hard X ray

Figure 6 shows the energy response of ZnO:Ga crystal to hard X ray, the energy range of the X ray fluorescence analysis device used here was 10–100 keV. The smooth line in the figure represents the fitting curve of the experimental data. We can draw a conclusion from it that the energy response to the X ray (>40 keV) is flat. On the other hand, Fig. 6

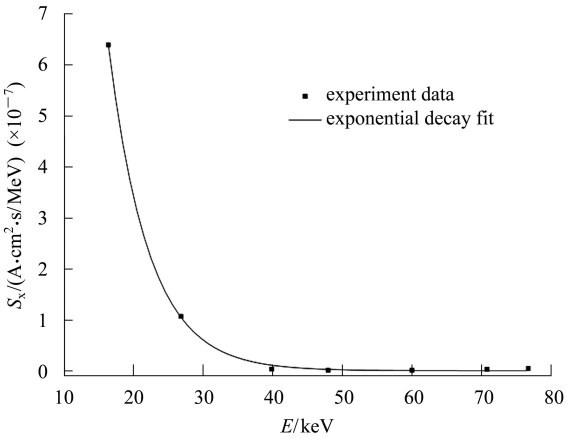

Fig. 6. The energy response of ZnO:Ga crystal to hard X ray.

donates the relations of incident energy (E) and the detection efficiency of the detecting system which

consisted of ZnO:Ga crystal and photomultiplier (S_X). The full-energy peak of the photon on the energy spectrum is defined as the peak formed in the detector where the total incident energy deposited, in which the photoelectric absorption is the highest contribution. The higher the energy, the smaller the photoelectric acting section except the absorption edge, the less the relative light output.

4 Proton excitation

The experiments were finished in the HI-13 tandem accelerator at the Chinese Institute of Atomic Energy, which can output a 4–26 MeV proton stream, with two modes: the pulsed proton and the constant current proton. The response curve of the ZnO:Ga crystal to proton was obtained successfully excited by the pulsed proton, which reflects the characteristic of the pulsed proton source. Fig. 7 shows the response of ZnO:Ga crystal to the 7 MeV pulsed proton. It was found that the signal impulses were periodic, and the periodic time was in accordance with the frequency multiplication of the proton source. When the proton interacts with the material of the sealing container, gamma radiation is emitted by (p, γ) reaction, gamma ray interacts with the ZnO:Ga crystal, then a light pulse is generated through a photoemission and Compton effect. This phenomenon can be seen in Fig. 7, where there is a short peak superposed on the rising edge of the negative impulse waveform. The oscillating peaks on the following edge are caused by an electronics effect, which is quite popular in the fast signal detection.

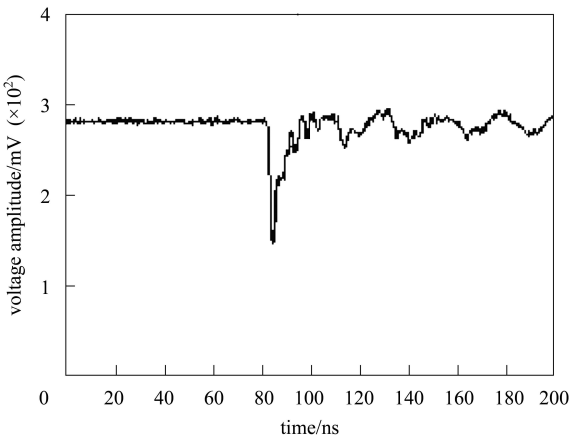

Fig. 7. The response of the ZnO:Ga crystal to 7 MeV pulsed proton.

By changing the energy of the constant current proton, the relationship between the luminescence intensity of ZnO:Ga crystal and the energy of the proton were obtained in Fig. 8. The smooth line is the exponential decay fitting curve. The thickness of the ZnO:Ga crystal used in the experiment was 300 μm, which can deposit 8 MeV proton completely. The bigger the energy is, the wider the range is, and the less the amplitude of the deposited energy in the ZnO:Ga crystal is. This result corresponds to that in Fig. 8, in which by increasing the energy of proton, the luminescence intensity is increased quickly also, but it will be increased a bit when the proton energy is greater than 8 MeV.

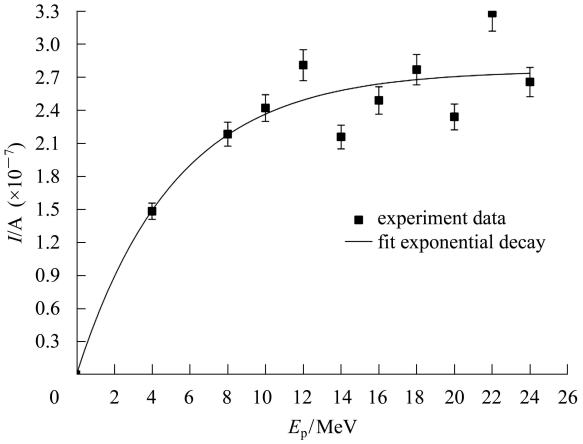

Fig. 8. The energy response of ZnO:Ga crystal to proton.

For improving the measurement accuracy, the beam must be adjusted before each test through fluorescent target in order to keep an effective interacting section and stable beam intensity. The crystal performance was affected greatly by the great dosage and long time irradiation of the proton. It must be noted that, after the proton irradiation, the ZnO:Ga crystal was activated intensively because of the (p, γ) reaction, thus the light intensity decreased to 90 percent. This phenomenon had been analyzed qualitatively in Ref. [7]. The radiation effect of gamma ray and laser on the ZnO:Ga crystal is completely penetrative, but the projective free path of the protons as other heavy charged particles in the crystal is much less than the photons. The charge collection of the heavy charged particles is different from the equalizing ionization of the photon in the whole sensitive region, the energy is generally higher, and the plasma effect comes into being. The high density of ionized plasma formed in the tracks of these charged particles caused the total destruction of the lattice. High energy proton irradiation can lead to the formation of thermal wedges with high temperatures in the lattice, thus the luminescence center was destroyed. In such lattice regions, the linear and volume defects, clusters,

etc., were formed, which play the role as leakage sites [8]. This results in the decreasing of the output energy of light, the shift of the central wavelength, and some changes in absorption and transmission spectra [9, 10].

5 Luminescence efficiency

The luminescence efficiency of the 200 μm ZnO:Ga crystal had been tested on the standard cobalt device in the Northwest Institute of Nuclear Technology. The intensity of radioactivity of the cobalt source was 1 Curie. Scintillation light was emitted when the energy of the gamma ray deposited in the scintillator, it transformed into electric current through a photomultiplier. The background current is measured by masking the photomultiplier with black paper. The light intensity of ZnO:Ga crystal, ST401, and BC418 had been measured at the same conditions. The results show that the response of ZnO:Ga crystal to the gamma ray was dearly better than that of the other scintillators, the output light intensity is 24 times larger than that of the 200 μm ST401 and 70 times than that of the 50 μm BC418. It is also found that the output light of the un-annealed ZnO:Ga crystal is 27 times lower than that of the annealed crystal, for annealing process can improve the quality of the crystal. The luminescence efficiency of ZnO:Ga crystal is higher than that of the anthracene. The number of γ rays incident in ZnO:Ga crystal has an inverse square proportion to the distance between the detector and the gamma source, when changing the distance, the illuminant intensity of ZnO:Ga crystal changes correspondingly.

6 Conclusion

ZnO:Ga crystal is a new type of inorganic scintillator, it has an excellent luminescence performance. The fast response to laser, X ray, and single α particle is sub-nanosecond. In addition, the scintillation efficiency of the ZnO:Ga crystal to the gamma ray is much better than that of the familiar ST401 organic scintillator.

The success in measuring the single α particle indicates that ZnO:Ga crystal can be used as an incidental alpha particle detector applied in a deuterium–tritium neutron generator, while further investigation is necessary. Moreover, the energy response to the X ray (>40 keV) is flat. The logarithm relationship of the energy response of ZnO:Ga crystal to X ray implies that it would be a new method for hard X ray energy spectrum detection. It was used successfully to measure the pulsed proton signal which reflects the characteristics of the proton source. This means that the ZnO:Ga crystal could be used in the detection system of the recoiled proton induced by pulsed neutron. But it could not be applied in the constant current proton measuring, because great dosage and long time irradiation of heavy particles can create radiation damage, which cause crystal structure and its optical property to be changed, thus the luminescent intensity is decreased intensely. Furthermore, it is most important to investigate experimentally the response of ZnO:Ga crystal to the different radiation particles, to study the luminescence center and the energy transfer mechanism, for expanding its potential use.

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