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Correlated analysis of 2 MeV proton-induced radiation damage in CdZnTe crystals using photoluminescence and thermally stimulated current techniques

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ABSTRACT

Radiation damage induced by 2 MeV protons in CdZnTe crystals has been studied by means of photoluminescence (PL) and thermally stimulated current (TSC) techniques. A notable quenching of PL intensity is observed in the regions irradiated with a fluence of 6×10^{13} p/cm^2, suggesting the increase of non-radiative recombination centers. Moreover, the intensity of emission peak $D_{complex}$ centered at 1.48 eV dominates in the PL spectrum obtained from irradiated regions, ascribed to the increase of interstitial dislocation loops and A centers. The intensity of TSC spectra in irradiated regions decreases compared to the virgin regions, resulting from the charge collection inefficiency caused by proton-induced recombination centers. By comparing the intensity of identified traps obtained from numerical fitting using simultaneous multiple peak analysis (SIMPA) method, it suggests that proton irradiation under such dose can introduce high density of dislocation and A-centers in CdZnTe crystals, consistent with PL results.

1. Introduction

CdZnTe (CZT) has been considered as one of the most promising materials in fabricating X-ray, gamma-ray and charged particle detectors [1], owing to its superior device performance at room temperature [2]. Since MeV charged particles have short ranges in semiconductors, usually several tens of μm, the preferred single charge carrier collection [3] widely used in hard X-ray and gamma-ray detection [4,5] can be easily achieved without complicated electrode geometry, which significantly improve the detector performance. More recently, an excellent energy resolution of about 1% has been achieved by detecting 2 MeV protons using commercial planar CdZnTe detectors [6].

The radiation detectors usually work in a harsh radiation environment, and thus a full knowledge of radiation damage is crucial for their application in various fields as well as to advance the radiation hardening techniques. Previous studies on charged particles-induced radiation damage in CdTe and CdZnTe detectors are mainly focused on its influence on the device performance [7–9]. For example, Zanarini et al. [10] investigated the damage induced by 2 MeV protons in CdZnTe and CdTe radiation detectors, which shows CdZnTe detectors suffer from performance degradation after a dose of 2.1–2.6 $\times 10^{11}$ p/cm^2, about an order of magnitude higher than that of CdTe detectors. Fraboni et al. [11] further suggests that the difference in stoichiometric ratio may account for the different radiation hardness between CdTe and CdZnTe detectors. Results of photo-induced current transient spectroscopy indicate that the trap labeled *Z* with an activation energy of 0.47–0.52 eV and the midgap trap labeled H1 with an activation energy of 0.79–0.82 eV may be responsible for electron trapping [12,13]. Further, time- and thermal-recovery processes of CdTe:Cl detectors suffering from radiation damage induced by protons and neutrons were investigated [14]. Hull et al. [15] and Eisen et al. [16] studied the effect of radiation damage induced by high-energy protons (∼200 MeV) on the spectroscopic characteristic of CdZnTe detectors. Though progress has been made on the influence of ion-induced radiation damage on the device performance of CdZnTe detectors [17], knowledge is still lacking on its microscopic electri-

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cal origins, in particular, the type of electrical active defects. Since the device performance strongly depends on these electrical active defects, it is therefore of importance to identify and, if possible, control the defects induced by impinging ions in CdZnTe crystals.

The main scope of this work is to study the defects responsible for the device performance degradation induced by 2 MeV protons in CdZnTe single crystals. Photoluminescence (PL) measurement was carried out to obtain information of structural changes in proton-irradiated regions. Thermally stimulated current (TSC) technology was performed to investigate the variation of trap levels in the bandgap, which are associated with electrical active states in the crystal lattice.

2. Experimental

A $7 \times 7 \times 2\ mm^3$ detector-grade $Cd_{0.9}Zn_{0.1}Te$ single crystal grown by the modified vertical Bridgman method in Imdetek was used in the experiments. Radiation damage was introduced using 2 MeV protons on the nuclear microprobe facility in the Institute of Modern Physics at Fudan University [18]. In this study, the CdZnTe sample was processed following the surface treatment procedure described in Ref. [6] but was not coated with Au electrode. Then, the crystal was exposed to broad-beam protons, in which the collimated beam spot was kept to be about $2 \times 2\ mm^2$ in area with a beam current of about 2 nA. Two regions, spaced 1 mm apart, were irradiated for 20 min and 5 min by the incident beam (Fig. 1(b)), with fluences of about $6 \times 10^{13}\ p/cm^2$ and $1.5 \times 10^{13}\ p/cm^2$, respectively. Device performance of CdZnTe detectors irradiated under similar fluence of 2 MeV protons was proved to deteriorate severely in previous studies [19]. Subsequently, PL measurements were performed on the irradiated regions to trace the fingerprint of radiation damage-induced changes in CdZnTe crystals. The CZT sample was attached on a cold finger in a liquid-helium cooled closed-loop cryostat. A 20 mW argon ion laser with a wavelength of 488 nm was used to excite the surface at 10 K. A Triax 550 tri-grating monochromator possessing a spectral resolution better than 0.3 nm was employed to collect the emissions and thus the PL spectrum was obtained. In order to acquire the difference of luminescence properties between irradiated and virgin regions, PL measurements were carried out linearly over the damaged regions point by point with a spacing of about 500 μm, as shown in Fig. 1(a). Following PL tests, three circular Au electrodes, with a diameter of 1 mm and being 1.5 mm spaced apart, were electrolessly deposited on irradiated and virgin regions, respectively, as shown in Fig. 1(b). Thermally stimulated current (TSC) technique was utilized to study the concentration variation of point defects as well as corresponding energy level shifts. A halogen lamp was used to fill the defective states at 10 K. During TSC measurements, the CdZnTe detector worked under an applied bias of 10 V. Consequently, TSC spectra were obtained by collecting current peaks, which result from thermally stimulated detrapping of electrons and holes from trapped states in darkness in the process of temperature increase from 100 K to 315 K. Immediately after, dark current spectra was recorded in the same manner, based on which the deep defect (E_{DD}) energy levels were calculated by applying the Arrhenius plot.

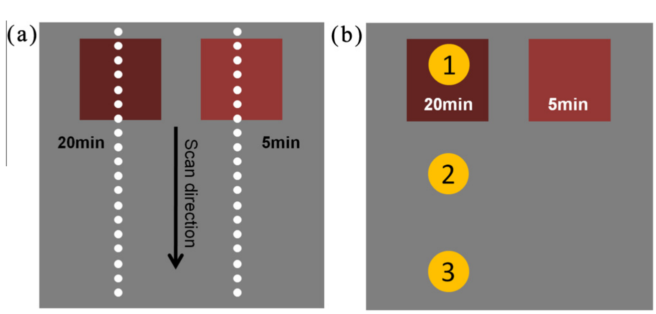

Fig. 1. Schematic diagram of (a) locations of 2 MeV proton-irradiated regions (square areas) on the CZT sample surface, with the linearly distributed points showing the positions where PL scanning was carried out and (b) electrode configuration (circular areas) for TSC measurements.

3. Results and discussion

3.1. PL measurement

Photoluminescence spectroscopy is a non-destructive technique to characterize crystal defects and provides abundant information to understand the recombination process in semiconductors. Fig. 2(a) and (b) show the distribution of PL spectrum across regions irradiated with fluence of $6 \times 10^{13}\ p/cm^2$ and $1.5 \times 10^{13}\ p/cm^2$, respectively. The uniformly distributed PL emission in unirradiated region suggests the high uniformity of crystallinity. However, the intensity of all the emission peaks in the PL spectrum decreases dramatically in irradiated regions. A comparison of Fig. 2(a) and (b) shows that higher dose of radiation damage leads to lower intensity of the emission peaks in PL spectrum, suggesting the increase of non-radiative defects in irradiated regions.

Fig. 2(c) shows the detail structure of PL spectrum from unirradiated and 5-min irradiated regions, where several emission peaks can be clearly identified, i.e., near-band-edge donor-bound exciton peak (D_0X) [1] at 1.649 eV, donor-acceptor pair (DAP) [20] recombination peak centered at 1.605 eV and its longitudinal optical phonon replicas (DAP-1LO) at 1.582 eV, a deep defect-related emission peak known as $D_{complex}$ [21] at 1.501 eV. The neutral acceptor-bound exciton (A_0,X) peak at 1.640 eV appears to be weak compared to D_0X. After irradiation of 2 MeV protons, the $D_{complex}$ peak dominates all the emission peaks, and the intensity of overall PL spectra decreases dramatically. The quenching of PL spectra suggests significant increase of non-radiative defects in irradiated regions, attributed to deep traps induced by radiation damage. Besides, all the PL peaks, including D_0X, $D_{complex}$ and DAP, are slightly shifted towards the low energies. The observed red-shift of PL spectra in irradiated regions may result from severe lattice distortions [22] or the modulation of band gap due to atom substitution [23,24], which is caused by radiation damage.

It was recognized that the defect-related deep emission peak $D_{complex}$ is caused by the overlapping of multiple emission peaks, and the identification of these emission peaks is helpful to understand the radiation-induced defects as well as their interaction with charge carriers. Therefore, Gaussian fitting is performed on $D_{complex}$ peak, and two peaks, namely D1 centered at 1.494–1.508 eV and D2 centered at 1.465–1.471 eV, are identified. The emission peak D1 is usually attributed to Cd-vacancy related complex called A-center [25], $[V_{Cd} - In_{Cd}]$. The luminescence of emission peak D2 caused by the recombination of exciton bound to dislocation related defects was also identified by Dean et al. [26]. Seto et al. [27] and Hildbrandt et al. [28] further confirmed that the intensity of this peak is strongly related to the dislocation density. Taylor et al. [29] found that Cd vacancies are stable at room temperature while Cd interstitials are not stable. Consequently, most of Cd vacancies induced by irradiation will anneal below 300 K due to interstitial migrations, and thus its concentration is relatively stable. Considering that the peak A_0X in CdZnTe crystal is attributed to an acceptor involving a Cd vacancy, the intensity of peak D2 with respect to that of peak A_0X can be used to evaluate the relative concentration fluctuation of corresponding defects regardless of the overall radiative efficiency change caused by