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Use of Sub-bandgap Illumination to Improve Radiation Detector Resolution of CdZnTe

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Abstract

The performance of $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ (CZT) materials as a room temperature gamma/X-ray radiation detector continues to improve in terms of material quality and detector design. In our prior publications, we investigated the use of multiple wavelengths of light (in the visible and infrared (IR)) to target charge carriers at various trap energies and physical positions throughout crystals. Light exposure significantly alters the charge mobility and improves carrier collection at the anode contact. This study presents an investigation of material performance as a radiation detector during such illumination. The decrease in charge trapping and increase in charge collection due to a higher probability of free electron release from traps contributed to an increase in the resolution-based performance of the detector through controlled illumination. We investigated the performance improvement of CZT crystals with previously known levels of intrinsic defects and secondary phases (SP), at various voltages, light-emitting diode (LED) light wavelengths and shaping times. Although our setup was clearly not optimized for radiation detector performance, it demonstrated substantial resolution improvements (based on the full width half maximum (FWHM) using the 662-keV gamma rays from ^{137}Cs upon illumination with 950 nm light) of 16 to 38% in comparison to un-illuminated CZT under similar conditions. This manuscript includes discussion of the

electro-optic behavior and its effect on performance. Additional testing and fabrication of a detector that incorporates such LED light optimization could lead to improved performance with existing detector-grade materials.

Introduction

CdZnTe or CZT can be used as an X-ray spectrometer as well as for X-ray imaging in the medical and aerospace industry. The development, fabrication and spectrometer performance of CZT has significantly improved over the last 20 years with performance benefits that under certain conditions begin to approach that of commercial Si and Ge detectors.^{1,2,3,4,5} One performance gap was narrowed through a better knowledge of internal electric field distribution by focusing on charge traps and reducing charge collection times for enhancing gamma interaction.^{6,7,8,9} As previously shown, the internal electric field of CZT can be altered through post growth manipulation using a variety of mechanisms.^{10,11,12} One of the least invasive methods of post-growth manipulation is the use of superficial illumination to interact with the embedded defects located in mid-gap energy states. The wavelength of illumination acts on the defect density in the mid-gap states to alleviate traps, induce in some cases, an electro-optic effect, and significantly increasing the carrier collection.^{10,13,14,15,16} An electro-optic effect with CdZnTe can be examined using a Pockels-like cell. Recently, the electro-optic effect was used to calibrate nuclear reactor pulses.¹⁷ During Pockels, near-infrared (NIR) radiation interacts with charge carriers within the crystal and causes a polarization of the internal electric field toward the cathode (negative).¹⁸

The internal electric field of CZT has been examined with materials that either have or possess the potential of high spectral resolution. We have previously investigated the influence of applying external light-emitting diode (LED) illumination (at an optimal power) to show visual evidence of the liberation of charge carriers that are trapped in low energy mid-gap states and in higher energy low gap states through Pockels imaging.¹⁸ One possible explanation for the enhanced electric field polarization includes the formation of a soliton resonance intensity, which is a self-focusing process that occurs in illuminated and electrically-biased CZT crystals.¹⁹ This room-temperature-based electro-optic behavior has been observed in materials that have electrons and holes as charge carriers, such as CZT and InP:Fe.²⁰ It involves the excitement of optically-sensitive electron traps and holes when the wavelength of background illumination is 1300 nm or less. [According to Schwartz, the

excitation of thermally-sensitive holes can occur separately and the level of excitation can even be controlled when the illumination wavelength is increased to 1500 nm.]¹⁹ For CZT, this type of extreme electro-optic performance as noted by Shwartz *et al.* (2006)²¹ and Schwartz *et al.* (2002)¹⁹ is limited to material that has been doped at the $\sim 10^{17} \text{ cm}^{-3}$ level and this self-focusing behavior does not occur in undoped material. Schwartz and colleagues (2002)¹⁹ have shown that in the absence of an applied electric field, the path of a circular 15- μm diameter light beam passing (or propagating) through a CZT wafer will increase in diameter by roughly three-fold indicating that there is a natural divergence within the crystal. Under bias, light beams can be focused to create a soliton in doped CZT. Schwartz and colleagues¹⁹ determined that there is a linear relationship between soliton intensity (self-focusing behavior for a fixed wavelength and entrance beam size) and illumination background intensity (in mW cm^{-2}) and that this linear relationship extends for more than four orders of magnitude.

Shwartz and researchers (2009)¹⁴ note that such electro-optic behavior is not limited to the surface of the CZT and that they are clearly found within the bulk of the material, which is expected given its low absorption of the sub-bandgap light. Solitons are not the only observations that involve electro-optic response behavior upon illumination in doped CZT. Sub-bandgap illumination of CZT “prepares” a more polar medium which can liberate deep traps but not substantially influence thermal processes that would otherwise interfere with the ionization. Illumination changes the lattice of the CZT, which causes the displacement of ions within the crystal lattice. Sub-bandgap illumination can also cause reversible changes in the bandgap (by 70 meV) and the refractive index of CZT.²² Charge carriers that move within the new electric field become separated and are later re-trapped. This type of polarization behavior with electro-optically stimulated CZT should influence the detection of incident light from radiation but the question is how? How does sub-bandgap illumination wavelength influence the signal process in a biased planar (doped) CZT detector? Zha *et al.* (2014) presented theory to explain sub-bandgap light enhancement on CZT radiation detector performance that uses a deep trap model, which supports an increase in the probability of deep trap releases of electrons.²³

Our presentation will present data on CZT performance. In this manuscript, we investigate a radiation detector performance enhancement of the internal electric field of CZT using illumination. The geometry used in our study favored a large degree of high capacitance behavior and therefore high electronic noise. This research

communication will report values that were far from optimal in terms of energy resolution. Prior work showed strong improvements in the electric field with these materials after sub-bandgap illumination using CZT detectors of known SP composition and radiation spectrometer performance.^{24,25} Improvements in detector performance with two of these CZT materials will be demonstrated through the use of energy resolution measurements. There is some overlap between the semiconductor radiation detector and the electro-optics research communities in terms of the impact of sub-bandgap light on the electrical behavior of CZT. This presentation explores the possible overlap between these research fields as it explores the possible explanations for the polarization response of CZT.

Experimental

The CZT crystals studied here were grown using either travelling heater method (THM) from Redlen Technologies (Victoria, B.C., Canada) or by modified Bridgman method from Yinnel Tech (YT, South Bend, IN) with an incorporation of 10% Zn shown previously by Triboulet *et al.* (1985).²⁶ The crystal from Redlen is referred to as “Redlen 64039B” and has the dimensions of $9.93 \times 10 \times 4.5 \text{ mm}^3$. A second crystal from YT is referred to as “YT 3-7-8” and has the dimensions of $12.1 \times 11.3 \times 6.48 \text{ mm}^3$. The two crystals were polished down to a fine polish with $0.05\text{-}\mu\text{m}$ alumina using standard techniques. Data on the resistivity, secondary phase composition/distributions, X-ray topography and detector performance of these materials were reported previously in more detail.^{18,27,28} Redlen 64039B was post-growth annealed and had a very low SP distribution relative to YT 3-7-8 (based on infrared (IR) transmission imaging) although these materials have been regarded as detector grade based on the radiation detector performance. Work by Awadella and co-researchers (2010) showed that this material possessed “vestiges” of SP in the topography that were not evident in IR transmission imaging studies. Regarding detector performance, YT 3-7-5 had an energy resolution of 1.7 % for the ^{137}Cs 662 keV gamma ray (bias of 1000 V and shaping time 0.3 μS), a bulk resistivity of $4.0 \times 10^{10} \Omega \text{ cm}$, an electron-mobility lifetime of $0.0025(\pm 5\%) \text{ cm}^2 \text{ V}^{-1}$, and the X-ray topographs for this CZT 3-7-8 material showed that it had more SP than the Redlen 64039B material. The Redlen 64039B material had a bulk resistivity of $7.3 \times 10^{10} \Omega \text{ cm}$ and an electron-mobility lifetime of $0.0068(\pm 5\%) \text{ cm}^2 \text{ V}^{-1}$. The Redlen 64039B had an energy resolution of 1.6 % for

the ^{137}Cs 662 keV gamma ray (bias of 1600 V and shaping time 0.5 μS). In the current study, the two CZT crystals were prepared with gold (Au) contacts that were sputtered for 60 sec. onto the crystals in a hemispherical geometry. Additionally, electrode-less Au was drop cast onto the four sides of each crystal and dried before measuring performance.

A Pockels-like setup as described previously by Groza *et al.* (2010) was used with a slight modification of using the LED light directly above the crystal.⁶ A LED was used as the source lamp at the near IR (950 and 1000 nm). Prior to making the energy resolution measurements, the optical power was varied at each wavelength to determine the optimal performance for each crystal. Using a 950 nm bandpass filter, the optimal performance was achieved at 120 μW (as measured). Using a 1000-nm bandpass filter, the optimal performance was achieved at 46 μW . The Au contact absorbed 80 to 85% of the optical power. This decreased optical powers on the surface of the CZT to: 18.96 μW of power at 950-nm light bandpass filter (Redlen 64039B), 7.37 μW at 1000-nm light with a bandpass filter (Redlen 64039B), 19.80 μW at 950-nm light with a 950-nm bandpass filter plus OD filter 1.0 (YT 3-7-8), and 14.38 μW of power with 1000-nm light and a 1000-nm bandpass filter plus OD filter 0.5 (YT 3-7-8). The illumination from the LED was directed parallel to the applied electric field onto the top face of the crystal—with the Cd-rich side facing up (**Fig. 1**).

Detector performance data were derived from pulse height discrimination measurements and calculated by dividing the full width half maximum (FWHM) of the full energy peak of the 662 keV gamma from ^{137}Cs . In our detector setup, an Amtek A250 CF charge sensitive preamplifier was used in conjunction with an ORTEC amplifier and a multi-channel analyzer (Canberra) model multiport II driven by Genie 2000, version 2.1. The bias was applied using a Keithley source meter and analyzed using SPIPTM software (from Image Metrology, Hørsholm, Denmark).

Results and Discussion

The focus of this study was to examine the relative improvement in detector energy resolution that occurs from the illumination of two CZT crystals with and without IR light (950 to 1000 nm) at various applied biases.

The geometry of the setup was not optimal for the purposes of detector evaluation but it allowed a preliminary investigation of the effects of illumination during radiation detection using a ^{137}Cs source.

The two CZT materials in this manuscript perform very well as radiation detectors under more optimized conditions. For example, the Redlen crystal had a FWHM of 1.6% for the 662-keV peak of ^{137}Cs , using a 1600-V bias, a 0.5- μs shaping time and single pixel geometry.²⁸ The YT 3-7-8 crystal had a FWHM of 1.7% for the 662 keV peak of ^{137}Cs using a bias of 1000 V, a 0.3- μs shaping time and a single pixel geometry.²⁷ Because our current study's focus was to examine the effect of sub-bandgap illumination on radiation detection, there were substantial changes in the detector performance setup that were required. **Figure 1** provides an image of the detector performance measurement setup. Prior work with these crystals in other geometries demonstrated that they had very high leakage currents (data not shown). Although measurements of detector resolution were possible, they were of poor quality—in the several tens of percent. To compensate for the high leakage currents, a layer of Au was added to four sides of the crystal and a hemi-spherical geometry was used with both crystals as noted in the methods. Due to high leakage currents with the Redlen crystal in particular, a virtual Frisch grid setup was also used.

Table I shows that sub-bandgap light illumination improved the energy resolution under all conditions. At a shaping time of 2.0 μs , there was better energy resolution at low voltage (500 V) for the 1000 nm illumination but in the absence of illumination, better resolution was obtained at 1000 V. At a lower shaping time (0.5 μs), a voltage of 1500 V and illumination at 1000 nm, the energy resolution was better than that at a higher shaping time of 2.0 μs at 1500 V. Lowering the illumination wavelength from 1000 nm to 950 nm produced the best total energy resolution improvements regardless of shaping time or voltage applied with. For the Redlen crystal, the resolution was better at an illumination of 950 nm as opposed to 1000 nm for the same bias and shaping time conditions. Higher biases did not always produce better resolution when that was the only parameter that was changed for the study of this detector. **Table I** also shows the percent improvement in resolution with illumination, which were the greatest when the crystal was illuminated at 950 nm as opposed to 1000 nm, regardless of the other setup parameters. Some example spectra for the data in **Table I** are shown in **Figs. 2, 3 and 4**.

Table II shows that NIR illumination (at the optimized power level as previously noted) always improved the energy resolution under all of the study conditions for the YT crystal. For illumination at 950 nm and 1000 nm and a constant shaping time of 2.0 μ s, higher voltages always produced better energy resolution. Use of lower shaping times at high biases produced better energy resolution regardless of illumination wavelength. Although work at low voltages did not produce the best energy resolution values with this detector, the measured overall resolution improvements (up to 38.2% at a low bias of 800 V) were the most substantial of all the test results for this crystal. **Table II** also shows the percent improvement in resolution with illumination, which was the greatest when the crystal was illuminated at 950 nm as opposed to 1000 nm. Some example spectra for the data in **Table II** are shown in **Fig. 5**.

Tables I and II show that the amount of energy resolution improvement was considerably greater at 950 nm than at 1000 nm with the exception of high voltage settings for the YT crystal at 2000 V, when there was little effect of wavelength on resolution improvement. The results from the YT testing differ from that with the Redlen crystal, which had fewer defects than that of the YT 3-7-8. According to transmission IR measurements with the two crystals, the Redlen crystal clearly has a lower number of SP relative to the YT^{27,28} and may also have relatively fewer types or numbers of traps that could be excited upon illumination. Additionally, the YT crystal which was more sensitive/responsive to the light may have had more trap “diversity” that were excitable with light, due to its greater number of defects and SP.

Growth with a Te-rich melt is nearly a prerequisite for the production of radiation spectrometer-grade CZT material, and such conditions are typical of modified vertical Bridgman growth (for example, as in Chaudhuri *et al.*, 2014) and other Bridgman growth methods.¹ Bridgman growth of CZT will ultimately produce some form of SP due to excess Te, although the size and quantity of the SP can vary.²⁹ High-quality spectrometer grade CZT will usually contain Te-rich SP. Too much Te-enrichment will often generate a material of poor spectrometer quality as studied by Bolotnikov and colleagues as well as many others.³⁰ However, low quantities/distributions of SP (originating from a Te-rich melt) are often associated with creating a high performance material as noted by Bolotnikov *et al.* (2007). Additionally, CZT that is grown with a Te-enriched melt can be greatly improved by post-growth annealing, which removes nearly all but a few vestiges of SP²⁸ as in

the case with the Redlen crystal and the annealing results in the formation of a high performance detector material. The Redlen material used in this study has been post-processed to reduce SP and has a different trap composition than the YT crystal, but its performance can be improved (energy resolution-wise) with light. Regardless, our present research findings demonstrate that light will enhance energy resolution, particularly when it has a power that is optimal for the specific detector as in this current study.

When the bias on the crystal was greatly increased (e.g., with 1500 and 2000 V with the YT and Redlen crystals, respectively in this study), additional noise degraded the signal quality in terms of resolution improvement. The decrease in resistivity upon illumination may be due to the freeing of traps that contribute to the free-carrier density; it could also provoke the decreased number of counts that are observed on the ROI for the 662-keV peak of ^{137}Cs , so there are some tradeoffs for the improvements in energy resolution.

Considerable energy resolution improvements were observed with illumination for the 662 keV peak. In prior studies, we performed studies where the optical power was varied from 0.0 to 62.6 μW with a constant bias of +350 V and have observed a ~20% decrease in the number of counts with increasing optical power on the LED (data obtained with a single pixel geometry at 950-nm illumination not shown). Illumination with 950-nm light results in a two orders-of-magnitude decrease in the bulk resistivity relative to that of the un-illuminated material depending of the defects concentration and light intensity.³¹ This effect was also closely observed by Shwartz and researchers (2009). Room temperature sub-bandgap illumination facilitates an increase in photo-induced free-carrier density which results in a higher capacitance (although there was some dependence on the light frequency).

Our study also did not directly determine the direct physical processes that helped improve the energy resolution but there are several possible explanations for this given previously discussed work on the effects of sub-bandgap light on the electrical behavior of biased CZT. The de-trapping of mid- and deep-level traps which should lead to greater charge collection is supported by a slow current decay after illumination during current transient measurements.³¹

We have shown that the sub-bandgap light frees trapped charge carriers, and we have used light to modify the internal electric field to a non-uniform state where the carrier density (or space charge region) is concentrated

near the negative contact (cathode). This charge collection results from holes that are released from the bulk that have migrated toward the cathode and now contribute to the overall signal. Additionally, removing trapped charge and decreasing potential electron-hole recombination sites in the mid gap region may provide for a clearer path for electron charge collection without substantial optically-induced excitation. Herein, the polarization-induced state generated through external illumination helped increase the energy resolution of the CZT in this study. Soliton behavior with CZT is not greatly subject to temperature-induced performance limitations,¹⁴ which (if active) would potentially help improve the thermal stability of CZT detector function at the upper end of most working temperatures³² in areas that require the use of radiation detection and characterization instrumentation.

Additional detector development would be required to have the flexibility to utilize 950-nm lighting during gamma-spectroscopy when resolution is most important but have the option of working without the illumination for less power consumption of the detector. Cola and Farella (2013)³³ examined the illumination of CdTe with light between 700 to 950 nm. Due to complete transmission of 950-nm light as opposed to the absorption of light at 850 nm, there were no directional effects on the illumination of CdTe by 950-nm light, which is consistent with our study with CZT.

Conclusions

In this study, the effect of illumination at 950 and 100 nm on the interaction of trapped charge carriers in CZT radiation detectors was investigated. The detector design was clearly not optimized for detection of radiation, but it allowed simultaneous sub-bandgap illumination during detector testing. However, improvements in energy resolution were clearly observed upon illumination. It is possible that the use of material of lesser quality may be realized with the illumination due to the enhancements that occur in the material performance as long as a more appropriate detector design can be utilized.

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Table I. Results of the illumination studies with Redlen 64039B crystal with hemispherical contacts and exposure to a ^{137}Cs source.

Shaping time (μs)	Light wave-length (nm)	Voltage (V)	FWHM with LED off (keV)	FWHM with LED on (keV)	FWHM Improvement (keV)	Resolution with LED off (%)	Resolution with LED on (%)	Resolution improvement ($\Delta\%$)	Total resolution improvement with illumination (%)
0.5	950	1500	24.1	16.5	7.5	3.7	2.7	1.0	27.1
2.0	950	500	28.6	19.4	9.1	4.3	2.9	1.4	31.9
0.5	1000	1500	26.3	20.6	5.7	4.0	3.1	0.9	21.6
2.0	1000	500	27.5	21.7	5.9	4.2	3.3	0.9	21.3
2.0	1000	750	27.3	21.9	5.4	4.1	3.3	0.8	19.7
2.0	1000	1000	26.5	22.2	4.3	4.0	3.4	0.7	16.2
2.0	1000	1500	29.9	24.0	5.9	4.5	3.6	0.9	19.6

Table II. Results of the illumination studies with YT 3-7-8 crystal with hemispherical contacts and exposure to a ^{137}Cs source.

Shaping time (μs)	Light wave-length (nm)	Voltage (V)	FWHM with LED off (keV)	FWHM with LED on (keV)	FWHM Improvement (keV)	Resolution with LED off (%)	Resolution with LED on (%)	Resolution improvement ($\Delta\%$)	Total resolution improvement with illumination (%)
3.0	950	800	32.6	20.2	12.5	4.9	3.1	1.9	38.2
2.0	950	1200	26.5	21.1	5.5	4.0	3.2	0.8	20.6
2.0	950	2000	22.9	17.6	5.4	3.5	2.7	0.8	23.3
3.0	1000	800	31.9	25.6	6.3	4.8	3.9	1.0	19.7
2.0	1000	2000	23.2	17.5	5.7	3.5	2.7	0.9	24.4

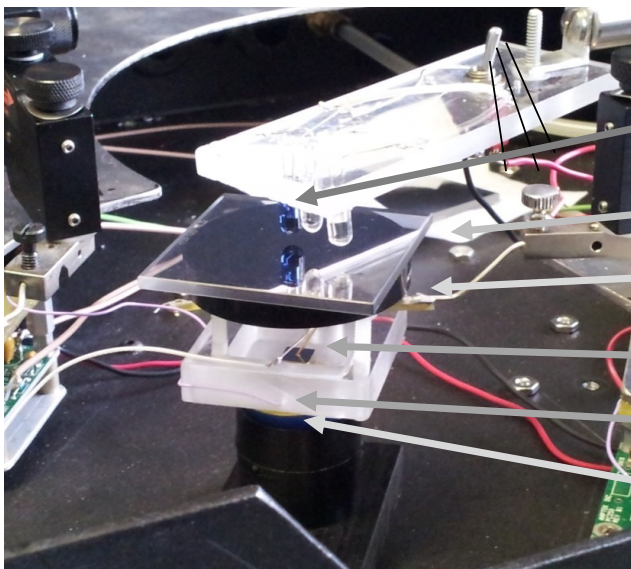


Fig. 1. The general setup of the illuminated detector studies with the YT 3-7-8 CZT crystal using hemispherical contact geometry. The studies with the YT crystal required optical density filters (0.5 and 1.0). The distance between the ^{137}Cs -137 source and the CZT was kept constant throughout the study. The same setup was used w

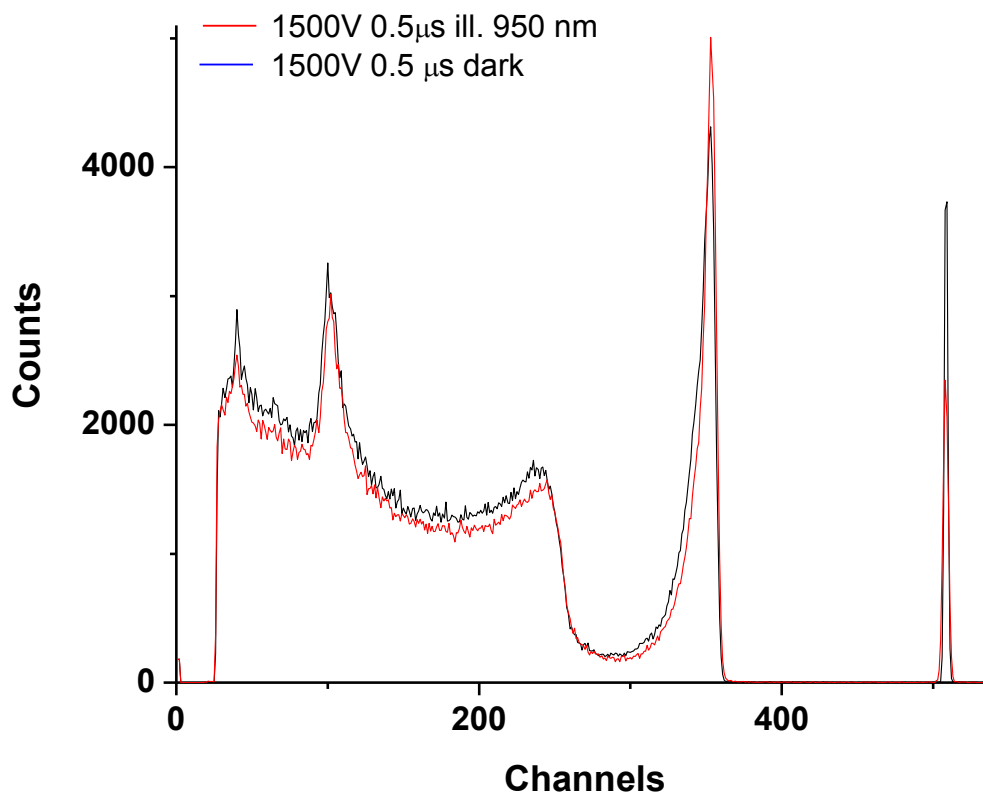


Fig. 2 ^{137}Cs with crystal 6403B in hemispheric configuration in dark and IR (950 nm) illumination.

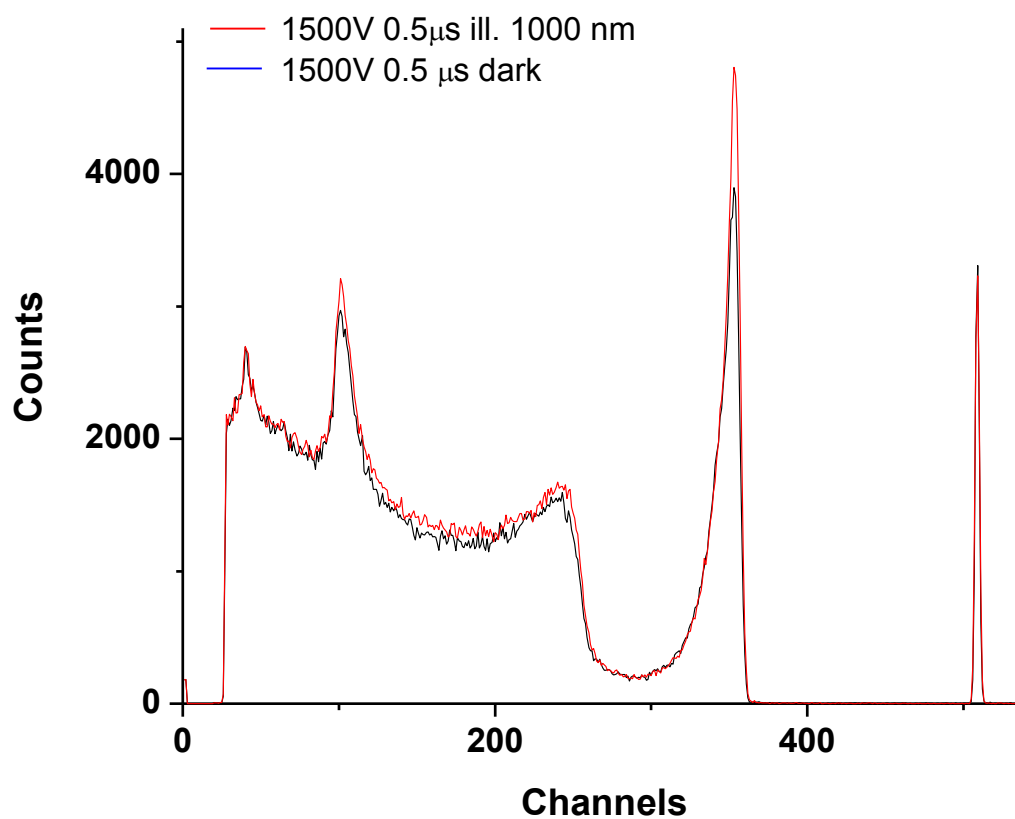


Fig. 3. ^{137}Cs gamma spectra obtained with CZT crystal 6403B in hemispheric configuration in dark and IR (1000 nm) illumination. With IR illumination, the energy resolution was improved from 4.0 to 3.1%.

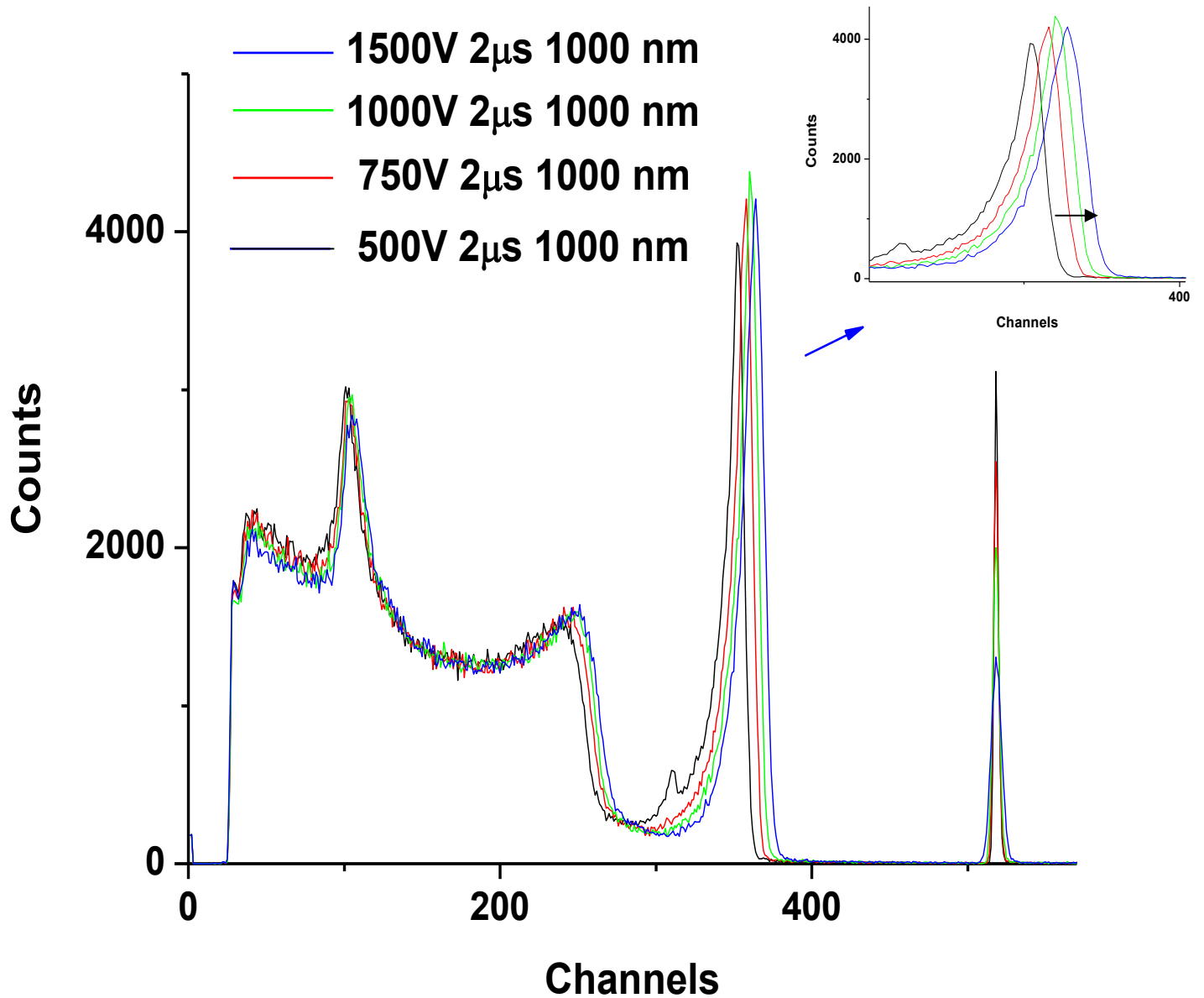


Fig. 4. Plot of gamma spectra for ^{137}Cs with crystal 64039B at several different voltages under anode illumination with IR 1000 nm. Some improvement in energy resolution occurred due to increased collection at higher bias as shown in the inset, with the horizontal arrow representing the effect of increasing bias on the energy resolution for the 662 keV peak.

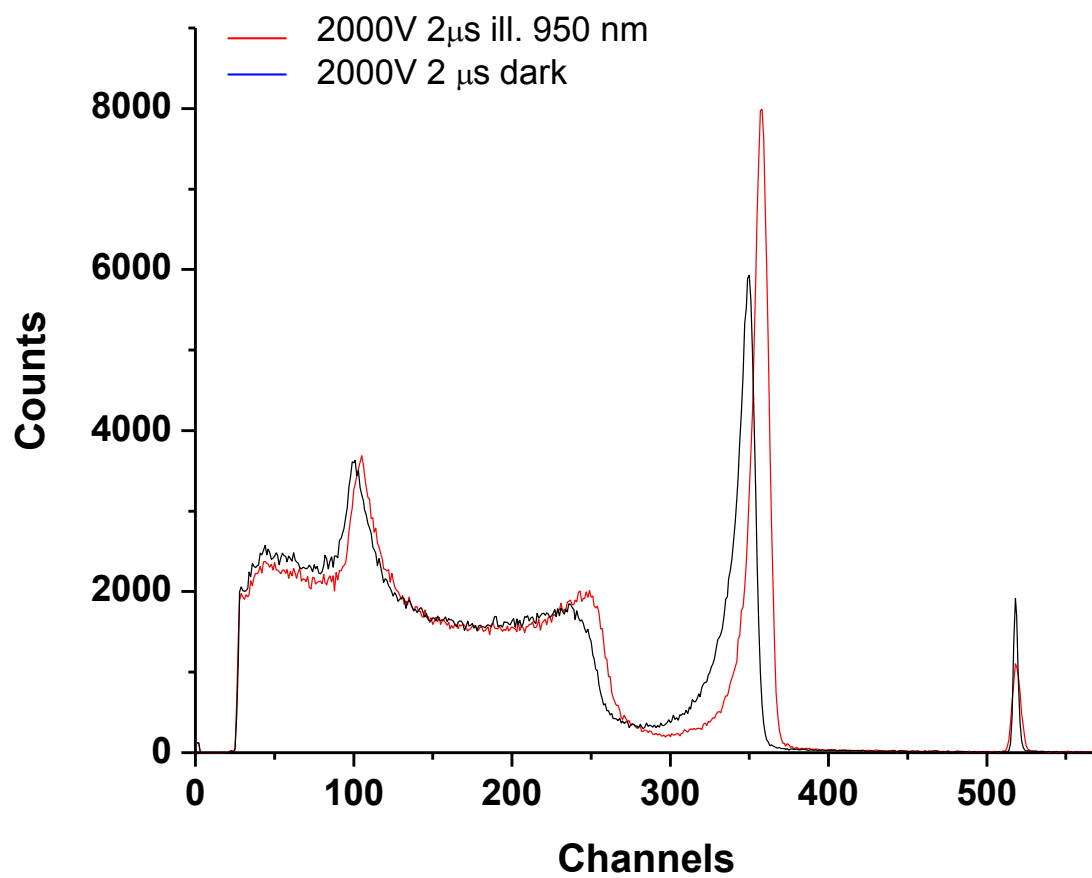


Fig. 5. ^{137}Cs using the YT 3-7-8 crystal (black spectrum) and 950 nm IR anode side illumination (red spectrum; with IR illumination) reveals better collection (higher channel) and better energy resolution (improvement from 3.5 to 2.7%).

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