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Two-crossed-polarizers based optical property modulation method for ionizing radiation detection for positron emission tomography

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Abstract

Recent work shows that Pockels effect and optics pump-probe measurement could be utilized as a novel method for 511 keV ionizing radiation photon detection for positron emission tomography (PET) which could potentially overcome the inherent physical limitation for coincidence time resolution of around 100 ps (Tao *et al* 2016 *Phys. Med. Biol.* **61** 7600–22). In this paper, we embrace this observation and introduce a two-crossed-polarizers based setup to achieve similar detection concept, which is a simpler and more compact setup with comparable ionizing radiation detection capability as the setup used in the previously proposed work. We evaluated the performance of our experimental setup with Lithium Niobate (LiNbO₃) and Cadmium Telluride (CdTe) detector crystals, and the desired properties of an ideal detector crystal were discussed. The modulation signal induced by 511 keV photons in both LiNbO₃ and CdTe can be detected with repeatable signal amplitude using two-crossed-polarizers based method, while CdTe could provide eight times higher detection sensitivity to 511 keV photons than LiNbO₃ under the same bias voltage, suggesting high effective Z number and high density properties of CdTe, as well as a shorter carrier lifetime and lower carrier mobility of LiNbO₃. In addition, the strength of modulation signal increased linearly with bias voltage before saturation. The modulation signal strength in LiNbO₃ continued to increase after 2000 V due to its high resistivity which could reduce the dark current in the detector, while the modulation signal of CdTe with low resistivity tended to be saturated at a bias voltage higher than 1200 V. Therefore, further increasing the bias voltage for detector crystals (especially for LiNbO₃) may enhance the modulation strength and improve the detection sensitivity for annihilation photons.

Keywords

Pockels effect; pump-probe measurement; Cadmium Telluride (CdTe); Lithium Niobate (LiNbO₃); time resolution; positron emission tomography (PET)

1. Introduction

Positron emission tomography (PET) is an imaging technique which uses positron-emitting radionuclide as a diagnostic probe in clinical medicine and physiological research with high sensitivity and specificity (Granov *et al* 2013, Pizzichemi 2016). Recent years the emergence of fast timing capability has become a new requirement mostly driven by the application of time-of-flight PET (TOF-PET) (Conti 2011, Lecoq *et al* 2014). The improvement of PET coincidence time resolution can bring many benefits over existing systems such as better trade-off between contrast recovery and noise (i.e. better signal-to-noise ratio) (Daube-Witherspoon *et al* 2010), image quality improvement and smaller lesion detection (Jakoby *et al* 2008, Karp *et al* 2008), shorter scan time with the same image quality (Conti 2011), dose reduction (Roberts *et al* 2005, Murray *et al* 2010), as well as overcoming inconsistent or missing data (Crespo *et al* 2007, Surti and Karp 2008). These advantageous characteristics could enable TOF-PET for normal clinical disease detection and some real-time imaging detections including guiding surgical treatments for diseases (Presotto *et al* 2015, Mehranian *et al* 2016).

As a consequence, a large amount of approaches to achieving better time resolution have been proposed including using new scintillation materials (Nemallapudi *et al* 2015), making a combination of different scintillators (Cates and Levin 2016), utilizing different sizes of SiPMs in combination with scintillators (Schaart *et al* 2010) and detecting Cerenkov light to acquire a better time resolution (Ariño-Estrada *et al* 2018). Some advances in coincidence time resolution have been achieved by these methods, but all scintillation based methods have an intrinsic time limitation placed by the process of scintillation which is estimated to be about 100 ps (Gundacker *et al* 2013) and the Cerenkov light based method is limited by the small amount of prompt photons produced which results in a very low detection efficiency (Miyata *et al* 2006). However, another promising method, optical property modulation based method (Tao *et al* 2016, 2017, Tao *et al* 2018), intended to utilize laser interference to achieve ionizing radiation photon detection with the potential to overcome drawbacks of previously proposed methods and showed possibilities to dramatically improve time resolution for PET, even to below 10 ps.

In optical property modulation based detection method (Tao *et al* 2016, 2018), researchers achieved ionizing radiation photon detection by utilizing the ultrafast phenomena of ionization charge carrier generation which could generate a detectable optical modulation signal. These ultrafast phenomena occur within a few picoseconds, which is significantly faster than the time resolution limitation of scintillation based detection method. By utilizing Pockels effect and optics pump-probe measurement (Gerd 2003), the group developed a spatial interference based setup with which the optical modulation signal induced by ionizing photons was successfully observed. However, it is well known that interference

system is a relatively complex and costly optical system composited with numbers of optical components. The visualization of spatial interference pattern has high requirements on instrumentation, such as a strict requirement on the surface parallelism and smoothness of the detector crystal sample. Besides, the whole setup would be relatively difficult to be integrated to a practical PET detector module due to the instability of interference and the sophisticated system structure. This motivates us to find a simpler and more practical optical arrangement with similar ionizing radiation photon detection capability.

In order to overcome these drawbacks of the spatial interference based setup, we propose a two-crossed-polarizers based method for 511 keV photon detection for PET, which only requires optical components that are inexpensive and easy-to-built. The approach discussed in this paper is also based on Pockels effect and optics pump-probe measurement. With this approach, we used a photodiode to detect the change of transmitted light intensity through an assembly containing two crossed polarizers, crystal detector (as Pockels cell) and lens. Ionization photons from a laser diode or from a radionuclide source can disturb the established electric field within the Pockels cell, thus altering the transmitted light intensity. Therefore, the detected signal in this paper is the change of light density. Using light transmission instead of using light reflecting from two opposite crystal surfaces (Tao *et al* 2016) avoids high requirements on crystal's surface condition. The simple arrangement of optical components makes our setup more compact and thus to be made into a practical PET detector module in the future.

In this paper, we focus on exploring the detection capability of two-crossed-polarizers based method and using the proposed method to detect the modulation of detector crystal's optical properties induced by an ionizing radiation source. We will also discuss the influence of material properties on detection results and sensitivity, and further talk about criteria for our future crystal material selection.

2. Material and method

2.1. Material selection for ionizing radiation detector

Detector crystals used in this work are LiNbO₃ and CdTe. Table 1 describes several characteristics of these two crystals including dimensions, refractive index, density, Pockels effect coefficient (i.e. linear electro-optic modulation coefficient) (Saleh and Teich 2007), effective Z number (Kaliman *et al* 2007), band gap energy (Wagner 2016), and resistivity.

The CdTe detector crystal is a $5 \times 5 \times 5$ mm³ cubic crystal which is chosen as a potential detector crystal due to its high effective Z number and high density that could provide satisfactory ionizing radiation photon interaction probability. The probe laser beam passes through two opposite surfaces which are well-polished and another two opposite surfaces are deposited with electrodes for supplying bias voltage.

The LiNbO₃ detector crystal is another $5 \times 5 \times 5$ mm³ cubic crystal which has comparably large Pockels effect coefficient and is widely used as ultra-fast optical switches in photonics devices using Pockels effect (Saleh and Teich 2007, Karp *et al* 2008). Compared to CdTe, the cost of LiNbO₃ is significantly lower. Much less complicated manufacturing process and

easily achieved good surface condition are another two advantages of LiNbO₃. Since the detection approach is based on Pockels effect, LiNbO₃, with a large Pockels effect coefficient and rapid modulation of optical properties under high energy photon excitation, is a desirable option.

2.2. Experimental setup

In this paper, a two-crossed-polarizers based method has been proposed as a new method for ionizing radiation detection for potential use in PET. The schematic diagram of the optical setup is shown in figure 1. Experimental layout of the detection system is shown in figure 2. The method uses the transmission of laser light (probe laser, WTD SOF-1550-D), which is initially linearly polarized and then illuminates the detector crystal. The collimated laser light transmits through the detector crystal sandwiched between two crossed polarizers (Thorlabs LPIREA200-C) which then is focused by a lens and reaches a photodiode (Thorlabs DET10N). The ionizing radiation photon interactions within the crystal can slightly change the established electric field. This disturbance of electric field modulates the polarized light phase directly, and subsequently changes the light transmission through the second polarizer, followed by a change of the output signal of photodiode. A lens is placed after the second polarizer to focus the transmitted light. Besides, light intensity after the second polarizer is the signal being monitored during the experiment. The two detector crystals are biased with a DC power supply (Hamamatsu C9727) and the entire optical setup is kept in a light-tight box.

2.3. Theoretical consideration

The mechanism underlying the ionization-induced optical property modulation is Pockels effect, which could modify the refractive index of the detector crystal due to the interaction between crystal and ionizing radiation photons. Both LiNbO₃ and CdTe were used as Pockels cells during our experiments. The modified refractive index of a Pockels cell can be expressed as equation (1)

$$n(E) \approx n_0 - \frac{1}{2}\gamma n_0^3 E \quad (1)$$

where $n(E)$ is the modified refractive index of the detector crystal with an applied electric field E . n_0 is the unmodified refractive index without applying an electric field, γ is the effective component of the Pockels effect coefficient and E is the magnitude of the applied electric field. Therefore, the refractive index of a Pockels cell is linearly dependent on the strength of the applied electric field (Esseling *et al* 2012).

For both crystals, the relationship between the light intensity passing through two crossed polarizers and the applied electric field can be expressed as equation (2) (Collett 2005),

$$I = I_0 \sin^2 \left(\frac{\pi n_0 \gamma d E}{\lambda} \right) \quad (2)$$

where I is the intensity of transmitted probe laser light, I_0 is the original intensity of laser or the maximum light intensity transmitted through parallel aligned polarizers, n_0 is the crystal's zero-bias refractive index, γ is the Pockels effect coefficient, d is the path length of light through detector crystal which is equal to the length of crystal, E is the magnitude of applied electric field, λ is the wavelength of transmitted light.

When ionizing radiation photon is absorbed in the detector crystal, charge carriers are generated. The application of a bias voltage across the Pockels cell can cause these free charges to drift toward their respective electrodes, producing a small internal electric field opposed to the externally applied electric field (established by DC voltage supply). In other words, the spatial movement of charge carriers alters the distribution of the electric field in the crystal. According to equation (1), the refractive index of detector crystal is changed due to the variation of electric field, which could modulate crystal's refractive index via Pockels effect and thus could change the polarization of light passing through crystal. According to equation (2), the intensity of transmitted light is dependent on the magnitude of electric field. Hence, a change in the bias voltage can alter the amount of light passing through the second polarizer, which can be measured with a photodiode. Consequently, ionizing interaction could alter crystal's refractive index and thus the intensity of light passing through the second polarizer could be changed, which can be detected by a photodiode. Therefore, monitoring the modulation of light intensity can help us observe the interaction between ionizing radiation photons and the detector crystal.

2.4. Measuring the resistivity of detector crystal

The resistivity of each crystal is an important factor which can influence the detection stability of the experimental setup. Therefore, we measured the current–voltage curve of each crystal. A programmable electrometer (Keithley 6517A) was used to detect the weak current flow in each crystal.

2.5. Testing the feasibility of optical setup

We initiated an experiment without ionizing radiation source aiming to test the feasibility of the two-crossed-polarizers based optical setup. We measured the dependence of transmitted light intensity on the bias voltage at 1550.00 nm probe laser wavelength. The best operating point of bias voltage was selected from these findings, as discussed in section 3.2.

2.6. Ionizing radiation experiment with laser diode

In order to ascertain the detection capability for refractive index change of our optical setup, we first used a laser diode as the ionizing radiation source, due to its controllable emission time and power. During our experiments, a 260 nm laser diode (Thorlabs LED260J) was used as the ionizing radiation source for LiNbO₃ and a 405 nm laser diode (US-Laser Inc.D405–120) was used for CdTe due to the difference of these two crystals' band gap energy.

2.6.1. Evaluation of the modulation signal—The change of optical signal level (output signal from the photodiode) was recorded by an oscilloscope (Tektronix AFG3022C) when each detector crystal was illuminated by laser diode light pulses. The oscilloscope was

also used to record the drive voltage of the laser diode. We then measured the dependence of modulation amplitude on bias voltage for two detector crystals, as discussed in section 3.3.2. In addition, we compared the optical modulation strength between two-crossed-polarizers based method and spatial interference based setup (Tao *et al* 2016) to evaluate the detection sensitivity of two methods, as discussed in section 4.

2.6.2. Evaluation of the charge carrier clearing time—Charge carrier clearing time is an important parameter as it both characterizes the timing performance of detector crystals and influences the timing property of our optical setup. In this paper, we used a free charge carrier counting method inspired by the pump and probe (P–P) method (Ling and Ajmera 1988) to calculate the charge carrier clearing time. The P–P method belongs to the class of transient method, which is based on free charge carrier absorption. Optical inducements, such as illuminating the crystal sample with a laser diode or with a radionuclide source, can generate free charge carriers in the detector crystal as long as the optical inducements' photon energy is higher than the band gap energy of crystal. The generation (as well as migration and recombination) of charge carriers could generate a variance in material's optical properties and thus we can measure the change of transmitted light intensity to achieve the measurement of charge carrier clearing time. Besides, this method gives a charge carrier clearing process directly and allows for detecting very short charge carrier life time (Laurentis and Irace 2014). The change of transmission light signal is given as equation (3),

$$v_L \approx V_L + v_0 e^{-t/\tau} \quad (3)$$

which shows that the transmission curve is an exponential function of t . v_L is the magnitude of exponentially decaying signal after removing optical inducements, V_L is the steady state signal level in the dark, v_0 is the starting value of exponentially decaying signal, t is the time variable, and τ is the decay constant indicating carrier clearing time.

In this work, we applied P–P method to measure the charge carrier clearing time. According to equation (1), ionizing radiation photons interact with detector crystal, producing free charge carriers which can modulate local electric field strength and the modulation alters the optical property of crystal (refractive index change in our paper). In other words, the detection of charge carriers is converted to the detection of refractive index modulation, which was measured by monitoring probe laser intensity, as discussed in section 2.2. Therefore, the charge carrier clearing time in the detector crystal can be estimated by analyzing the modulated signal caused by laser diode or radionuclide sources.

With the same method as described in section 2.6.1, pulses from the laser diode were cast onto the detector crystal and we recorded the change of signal magnitude by a photodiode. The same procedure was repeated for ten times for each crystal and all results were averaged to one final result for our further analysis. The frequency of the drive voltage was set to 1 Hz in order to allow charge carriers to recombine completely and to ensure the signal level is recovered to the original (dark) level again.

2.7. Ionizing radiation experiment with Na-22 radionuclide source

After using a laser diode as the ionization radiation source, we then observed the modulation signal induced by radionuclide sources.

A 100 μCi Na-22 source (Rocca and Riggi 2016) was used in experiments. The source was repeatedly placed 15 mm from LiNbO_3 and CdTe crystals and then removed. We observed and recorded the change of signal level between presence and absence of the source, in order to see how a radionuclide source modulates the signal level. The selected data point for each measurement is the stable signal level observed at 5 min after placing or removing the source. This is because the signal did not change immediately during the transition between the delivery of source to the detector crystal and the removal of source. After the settling time, the signal level remained stable at a fixed value. Therefore, we waited 5 min for a stable signal level. Each data point was recorded as a histogram of the signal magnitude for 30 s. The value corresponding to the peak of the histogram is used as the magnitude of the stable signal level. All data points were taken consecutively in time.

3. Results

3.1. Resistivity of detector crystals

The current–voltage curves of LiNbO_3 and CdTe are shown in figure 3. According to current–voltage measurement and Ampere’s laws, we estimate that the resistivity for LiNbO_3 and CdTe are $30.5 \text{ G}\Omega \cdot \text{m}$ and $19.6 \text{ M}\Omega \cdot \text{m}$, respectively. The resistivity of LiNbO_3 is three orders of magnitude higher than that of CdTe, therefore LiNbO_3 has a much larger resistance with the same dimensions.

3.2. The feasibility of setup

According to equation (1), changing the magnitude of the external electric field may alter the refractive index of crystal due to Pockels effect. Therefore, according to equation (2), the intensity of light passing through two crossed polarizers is modulated as the electric field magnitude is adjusted, and then we can monitor the light modulation by monitoring the output signal level of photodiode.

For each crystal, the dependence of signal level on bias voltage at 1550 nm probe laser wavelength is shown in figure 4. The results demonstrate detectable Pockels effect in both LiNbO_3 and CdTe, and reveal the feasibility of our proposed method for detecting the modulation of crystal’s refractive index induced by ionizing radiation photon. The generation of charge carriers can modulate the internal electric field of crystal, thus could modulate the signal level. For our following experiments, we mainly focus on the modulation signal induced by crystal internal electric field disturbance.

Moreover, a larger modulation signal level of LiNbO_3 crystal is related to its larger Pockels effect coefficient. The longer oscillation period of LiNbO_3 crystal is related to its higher resistivity, as discussed in section 4.

3.3. Experimental results with laser diode as the ionization source

3.3.1. Modulation signal detection—Figures 5(a) and (b) show the modulation signals with a laser diode as the ionizing radiation source for LiNbO₃ and CdTe, respectively. The red line represents the drive voltage of laser diode, while the blue line indicates the changes in modulation signal level.

For LiNbO₃, the magnitude of optical modulation signal level goes up from 0 V to 3500 V. 3500 V bias voltage, the largest output of DC power supply, is chosen as the operating voltage. For CdTe, 1600 V bias voltage is chosen as its operating bias voltage, with which the optical modulation signal level is at its maximum value. In order to clearly show the shape of the optical modulation signal, the modulation signals of the two crystals are shown with different bias voltages and the optical modulation signal level is normalized to the signal level in dark. The selection of the optimal operating voltage for two crystals will also be discussed with more details in section 3.3.2.

Our findings indicate that the optical properties (refractive index in this paper) of LiNbO₃ and CdTe can be modulated by a laser diode as the ionizing radiation source, and our setup can be used to detect these relatively weak modulation signals as illustrated in figure 5. The reasons for the difference in the optical modulation signals of LiNbO₃ and CdTe will be discussed in section 4. Furthermore, a comparison is done between the modulation signals detected by the two-crossed-polarizers based method and by the optical interference based method (Tao *et al* 2016), which is discussed in detail in section 4.

3.3.2. Optimal operating voltage selection—To better illustrate the selection process of the optimal operating voltage, we define a new metric, the optical modulation strength, to evaluate the detection sensitivity of our optical setup. The quantity can be expressed as equation (4),

$$strength_{optical\ modulation} = Stable_{signal} - Max_{signal} \quad (4)$$

where $Strength_{optical\ modulation}$ is the optical modulation strength. $Stable_{signal}$ represents the stable signal level in dark (the baseline signal level in figure 5) and Max_{signal} is the point where the maximum modulation occurs (the lowest signal level point in figure 5). Bigger $Strength_{optical\ modulation}$ indicates a better detection sensitivity.

The dependence of optical modulation strength on bias voltage for LiNbO₃ and CdTe are shown in figure 6. In order to exclude the influence of different bias voltages and to clearly show the comparison between two crystal, we normalize the optical modulation strength at each testing bias voltage to the optical modulation strength at the optimal operating bias voltage, which corresponds to the largest modulation strength. From figure 6, we can conclude that the optical modulation strength is linearly proportional to bias voltage for both two crystals, but with different changing trend. Higher bias voltages applied to crystal can enhance the optical modulation strength. The amplitude of the modulation signal induced in LiNbO₃ goes up from 0 V to 3500 V, while the amplitude of the modulation signal induced in CdTe tends to be saturated at bias voltages greater than 1600 V. Further increasing the

bias voltages may only contribute a small increment to the magnitude of the modulation signal. Therefore, 3500 V is selected as the operating point for LiNbO₃ and 1600 V is selected as the operating point for CdTe. We will further discuss the influence of different bias voltage on the magnitude of optical modulation strength in detail in section 4.

3.3.3. Calculation of carrier clearing time—For LiNbO₃, the modulation signal induced by a laser diode pulse is presented in figure 7(a) which is also an inverted view of the modulation signal shown in figure 5(a) after subtracting the baseline signal level. Compared with equation (3), the remaining value in figure 7(a) corresponds to $v_L - V_L$. Figure 7(b) illustrates several data points selected from the decay edge of the modulation signal with y axis plotted on natural logarithm scale. By simply taking the natural logarithm of both sides in equation (3), we observe that the slope of the straight-line region corresponds to $(-1)/\tau$, as shown in equation (5)

$$\ln\left(\frac{v_L - V_L}{v_0}\right) \approx \frac{(-1)}{\tau}t. \quad (5)$$

Therefore, according to equation (5), the charge clearing time of LiNbO₃ crystal is estimated to be $\tau_{\text{LiNbO}_3} = (-1)/(-706.696.43) = 1.42\mu\text{s}$, while the charge clearing time of CdTe crystal is estimated to be $\tau_{\text{CdTe}} = (-1)/(-5912.8) = 169.12\mu\text{s}$, as shown in figure 8.

3.4. Experimental results with Na-22 as the ionization source

The modulation signals of LiNbO₃ and CdTe modulated by a Na-22 ionizing source are shown in figure 9. In order to ensure that the CdTe detector is not saturated, the data for CdTe were obtained under a bias voltage of 1200 V. To have a better comparison with CdTe, the data for LiNbO₃ were also obtained under 1200 V bias voltage.

As shown in figure 9, the output optical signal from both LiNbO₃ and CdTe crystals can be modulated by the presence of the Na-22 radionuclide source, due to a change in the refractive index of both detector crystals. However, we can see that there is eight times difference in the magnitude of modulation signal between LiNbO₃ and CdTe, which is discussed in section 4. A further comparison of the modulation signal level induced by Na-22 between two-crossed-polarizers based method and optical interference based method (Tao *et al* 2016) is also discussed in detail in section 4.

Moreover, the relationship between the optical modulation strength induced by Na-22 and the crystal bias voltage is shown in figure 10. An increased crystal bias voltage leads to a stronger optical modulation strength for Na-22 induced modulation signal. The modulation signal induced in CdTe tends to saturate at bias voltages larger than 1200 V, and thus the last three data points are not included in the linear fitting. However, the modulation signal induced in LiNbO₃ increases continuously with the increase of bias voltage from 0 V to 2000 V. We will discuss this point in section 4.

4. Discussion

4.1. Potential of two-crossed-polarizers based method for improving time resolution for PET

The two-crossed-polarizers based detection method depends on the Pockels effect, which is an instantaneous optical modulation effect. Therefore, the detection speed only depends on the creation speed of the ionization charges which is an ultrafast process (sub-picosecond scale to femtosecond scale). The time resolution of the two-crossed-polarizers based method is also limited by the detection sensitivity of our setup, which determines the number of free charge carriers needed before a detectable signal shows up. Some other factors, such as the timing properties of photodiode, oscilloscope and other devices involved will also influence the achievable timing resolution of our method. Therefore, even though the time constant observed in figures 5(a) and (b) is slow. The intrinsic physical limit for the time resolution of the proposed method is still expected to be at the sub-picosecond time scale. Possibilities exist to make use of ultrafast optical modulation phenomena to push the detection speed towards the intrinsic physical limit for timing. Therefore, the method is promising for dramatically improving PET time resolution.

Although the paper has described the studies with LiNbO₃ and CdTe to study the feasibility of ionizing radiation photon detection, this method does not rely on the two crystals. Other materials with larger Pockels effect coefficient (thus better detection sensitivity) will also be considered in our future studies.

4.2. Feasibility of setup

From figure 4, we can see that the change of the normalized optical signal level for LiNbO₃ is around 0.225 from 0 V to 2000 V while that variation for CdTe is around 0.033. Different performances in modulation signal level mainly result from different Pockels effect coefficient of each crystal. According to table 1, LiNbO₃ has a relatively larger Pockels effect coefficient than CdTe. Therefore, LiNbO₃ can generate a more obvious modulation in signal under the same bias voltage. It also indicates that a large Pockels effect coefficient can contribute to a better detection sensitivity and this characteristic should be taken into consideration for our future detector crystal selection.

We can also see from figure 4 that CdTe with low resistivity presents almost one complete oscillation cycle from 0 V to 2000 V, while LiNbO₃ with high resistivity presents less than three quarters of an oscillation cycle from 0 V to 2000 V. Therefore, the resistivity of the crystal could influence our detection results. We also notice that, when adjusting the bias voltage and keeping the output current of the power supply at a constant value, we did not observe obvious optical signal change. Therefore, the induced current in each crystal is a decisive factor for the detection results. The resistivity of each crystal affects the induced current in the crystal and thus will affect the final detection results. Crystals with larger resistivity could generate a smaller induced current. For our future crystal selection, it indicates that selecting a crystal with large resistivity can reduce the signal noise level (due to reduced dark current) and contribute to a better detection signal stability.

4.3. Analysis for the experiment results with laser diode

As shown in figures 5(a) and (b), the modulation signal induced by laser diode showed up with both LiNbO₃ and CdTe, but the charge carrier clearing time observed in LiNbO₃ is much shorter than that in CdTe. This distinction in charge clearing time is mainly due to the differences in charge carrier mobility, bias voltage and charge carrier lifetime between LiNbO₃ and CdTe. We first focus on the charge carrier mobility, the charge carrier mobility in CdTe is approximately 1000 cm² Vs⁻¹ (Bell 1977), as compared to less than 5 cm² Vs⁻¹ (Gerson and Kirchhoff 1986) in LiNbO₃. By taking bias voltage into consideration (LiNbO₃ at 3500 V and CdTe at 1600 V), we can estimate that the electron drift velocity in CdTe is more than 100 times larger than that in LiNbO₃. By analyzing the decay edge of the two induced signal, the charge carrier clearing time of LiNbO₃ is estimated to be around 1.42 μ s while that value of CdTe is approximately 100 times larger around 169 μ s, which matches the ratio between the electron drift velocities in the two crystals. On the other hand, the charge carrier lifetime in LiNbO₃ is reckoned to be around 0.1 ns to 1 ns (Gerson and Kirchhoff 1986) and the lifetime of CdTe is reported to be on the order of 0.2 μ s (Bell 1977). With the charge carrier lifetime in LiNbO₃ being 2–3 orders of magnitude shorter than that of CdTe, the charge carrier clearing time of LiNbO₃ is also much shorter than that of CdTe. Therefore, we can conclude that an ideal detector material requires high carrier mobility, and short carrier lifetime which indicates short detector dead-time, in order to provide the optimum detection efficiencies for measuring 511 keV photon interactions.

From figure 6, we can see that the modulation signal level is linearly dependent on bias voltage before saturation. The modulation signal induced in LiNbO₃ increases with bias voltage from 0 V to 3500 V while in CdTe the modulation signal tends to be saturated at bias voltages greater than 1600 V. Since the induced current determines the strength of optical modulation, the reason of saturation is also related to the induced current. For CdTe, when the bias voltage is lower than 1600 V, the induced current is a main limiting factor for signal magnitude increase, so we can increase the optical modulation strength by increasing bias voltage. The optical modulation signal magnitude tends to be saturated with a bias voltage higher than 1600 V, possibly because of the energy limitation of the laser diode pulses instead of the induced current. Since the induced current has reflected all absorbed laser photons under high bias voltage, the output energy of the laser diode is the main limitation for further improving signal magnitude. In other words, we can increase the optical modulation signal by increasing the output energy of laser diode after saturation.

4.4. Results with radionuclide source as ionizing radiation

We observe from figure 10 that LiNbO₃ crystal is not as sensitive as CdTe crystal for the detection of 511 keV photons under the same bias voltage. Different detection sensitivity of the two crystals is mainly due to their different properties, including Pockels effect coefficient, density and band gap energy. LiNbO₃ crystal has a greater Pockels effect coefficient which could produce a stronger modulation signal, but the density and the Z number of CdTe is much higher than LiNbO₃, which means CdTe has a stronger photon stopping ability enabling it to absorb more energy from ionizing radiation and improving its capability to interact with ionizing photons. The band gap energy of the crystal is indirectly related to the number of carriers generated by 511 keV photon interactions, and thus

influences the optical modulation strength. The lower the band gap is, the easier it is to generate more charge carriers with the same strength stimulus. Notably, CdTe has a lower band gap energy (1.5 keV for CdTe and 3.8 keV for LiNbO₃). Hence, it is expected that more carriers are produced in CdTe from 511 keV photons. In summary, the combination of these factors results in the different detection sensitivity of the two materials. A significantly larger number of ionization-induced charge carriers produced in CdTe increases the optical modulation strength compared to LiNbO₃.

Furthermore, we found that the baseline optical signal level tended to drift more severely in CdTe with low resistivity. Such observations are believed to be related to the large dark current induced in the CdTe crystal under the same bias voltage. The modulation signal of CdTe tends to be saturated at bias voltage higher than 1200 V, which is associated with the low resistivity (larger dark current) as well, while the signal with LiNbO₃ still goes up after 2000 V which is in correspondence with the results of the spatial interference based method (Tao *et al* 2017).

As described in section 2.7, there is 5 min waiting time for reading out the stable signal level after the equilibrium is established in LiNbO₃ and CdTe. However, this does not mean that the setup proposed is limited to five minutes readout delay, since the signal level starts to change as soon as the ionizing radiation source is placed close to the detector crystal which is theoretically an ultrafast process under 10 ps. Therefore, searching for a faster readout method would be a subject for our future study. Our ultimate goal would be to establish a setup (including developing an appropriate readout method) that is sensitive enough to measure the modulation signal from the ionization charges created by individual 511 keV photons with measured response time and variance both on the order of picosecond.

4.5. Comparison with interference based method in detection sensitivity

The detection sensitivity (based on CdTe) of our proposed setup and the interference based method (Tao *et al* 2016) is compared. Since the laser diode and the radionuclide source we use are similar with the interference based method, we could ignore the influence of the laser diode power and the radiation source flux on our results. The laser diode induced signal fluctuation magnitude is 0.05 (normalized) for our present work and 0.01 (normalized) for the interference pattern based method. The radionuclide source induced signal fluctuation magnitude is 0.08 (normalized) for our present work and 0.23 (normalized) for the interference pattern based method. With laser diode as the source of ionizing radiation, data revealed that the sensitivity of two-crossed-polarizers based setup is five times higher than the interference pattern based method. While, for Na-22 as the ionizing radiation source, the sensitivity of two-crossed-polarizers based method is about 1/3 of the sensitivity of spatial interference based method. Though our proposed setup is already able to detect the relatively weak and noisy modulated signal induced by a radionuclide source, our future work will mainly focus on the approaches to improving the detection sensitivity and reducing the noise level for the two-crossed-polarizers based setup. Finally, we aim to accomplish single 511 keV photon detection and achieve detection time resolution less than 10 ps.

5. Conclusions and future work

5.1. Conclusions

In this paper, we borrowed the concept of Pockels effect and optical pump-probe measurement to propose a new optical setup structure, which was named as two-crossed-polarizers based method. We tested the performance of our proposed method with LiNbO₃ and CdTe, and pointed out the desired properties of an ideal detector crystal for this method.

Pockels effect combined with optical pump-probe measurement was successfully implemented for the detection of ionizing radiation photons. We have detected the modulation signal induced by a laser diode and by a radionuclide source (Na-22) as the ionizing radiation source using both LiNbO₃ and CdTe. When using a laser diode as the ionizing source, LiNbO₃ with a relatively short carrier lifetime and relatively slow carrier mobility has a shorter charge carriers clearing time than CdTe. When using Na-22 as the ionizing source, CdTe with relatively high density and high Z number is more sensitive to 511 keV photons than LiNbO₃. Besides, our data indicate that the amplitude of the optical modulation signal increases linearly with increasing bias voltage for both two crystals before saturation. Further increasing the bias voltage for detector crystal could hypothetically enhance the modulation strength and detection sensitivity of our method, especially for LiNbO₃.

In addition, a comparison between our work of two-crossed-polarizers based method and the previous work of interference based method reveals the advantages and disadvantages of our setup. Although our method in this paper cannot be as sensitive as the interference based setup for radionuclide source, the compact structure, easy-to-built optical setup and low requirements for crystal surface condition still provide great advantages and potential for its future application as a practical PET detector.

5.2. Future work

Our future work will focus on the improvements of the detection sensitivity of two-crossed-polarizers based optical method. We believe that further gains in the setup sensitivity and reduction in the noise level can get us closer to achieving single 511 keV photon detection with optimized time resolution.

With the hope to get a better detection sensitivity, we will research the magic angle (Tan *et al* 2005) of our two-crossed-polarizers based setup. The magic angle is a special working angle between two polarizers which could dramatically decrease the noise component of the signal with only insignificant loss of valid signal. In this case, the sensitivity of the setup could be improved significantly.

2D polarization selective IR pump-probe (2D IR) experiments (Kramer *et al* 2015) has been widely used to study ultrafast dynamics, including interaction of a range of molecular systems and reactions which involve the transfer or movement of molecules on a time scale of 10 fs to 1 ps. We plan to borrow the ultrafast detection concept from 2D IR technique to our two-crossed-polarizers based method. The combination of the two methods might

improve the sensitivity of our setup with the ultimate goal to achieve individual 511 keV photon detection and improve the time resolution of PET.

The wavelength of the probe laser is another important factor since the transmission efficiency of the probe laser will vary dramatically at different wavelengths, so we will apply both 850 nm for CdTe and 2 μm for LiNbO₃ as the probe laser wavelength. Since these chosen wavelengths are close to each crystal's band gap energy, it might enhance the optical modulation strength and thus improve the detection sensitivity of our setup. In addition, we will continue to explore other high Z number and high density detector crystals. CdTe and LiNbO₃ crystals with different electrode patterns will also be considered and researched.

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Figure 1.
Schematic diagram of two-crossed-polarizers based setup.

Figure 2.

Experimental layout of the detection setup with a laser diode (together with focusing lens) as the ionizing radiation source. A radionuclide source (Na-22) can be placed beside detector crystal replacing the laser diode to measure 511 keV ionizing radiation-induced modulation signal.

Figure 3.
Current–voltage curves for (a) LiNbO₃ and (b) CdTe.

Figure 4. Dependence of signal level on bias voltage without ionizing radiation photon for (a) LiNbO₃ and (b) CdTe.

Figure 5.

(a) Optical modulation signal induced by laser diode as ionization source for LiNbO_3 ; (b) optical modulation signal induced by laser diode as ionization source for CdTe . The modulation signal is consisted with a small modulated signal on top of a large stable signal in dark (i.e. DC component of signal without laser diode modulation).

Figure 6.

(a) LiNbO₃: the dependence of optical modulation strength on bias voltage; (b) CdTe: the dependence of optical modulation strength on bias voltage.

Figure 7.

LiNbO₃: (a) modulation signal induced by laser diode; (b) data points selected from the decay edge of pulse modulation signal induced by laser diode.

Figure 8.

CdTe: (a) modulation signal induced by laser diode; (b) data points selected from the decay edge of pulse modulation signal induced by laser diode.

Figure 9.

Modulation signals for (a) LiNbO₃ and (b) CdTe induced by Na-22. Red bars indicates the magnitude of modulation signal when Na-22 source is placed nearby detector crystals, whereas blue bars represent the magnitude of signal when source is removed.

Figure 10. Dependence of Na-22 induced optical modulation strength on crystal bias voltage for LiNbO₃ (blue line) and CdTe (red line).

Table 1.Comparison of several crystal properties between LiNbO₃ and CdTe.

Crystal	LiNbO ₃	CdTe
Dimensions	$5 \times 5 \times 5 \text{ mm}^3$	$5 \times 5 \times 5 \text{ mm}^3$
Refractive index	2.29	2.74
Density	4.3 g cm^{-3}	5.85 g cm^{-3}
Pockels effect coefficient	32 pm V^{-1}	6.8 pm V^{-1}
Effective Z number	34.6	50.2
Band gap energy	3.8 eV	1.5 eV
Resistivity	High	Low