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Article

Keywords:

Posted Date: August 9th, 2022

DOI: https://doi.org/10.21203/rs.3.rs-1936977/v1

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High-harmonic generation in polycrystalline CdTe nano-films via macroscopic investigations

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Bright high harmonics generation (HHG) in CMOS-compatible nano-films can provide new opportunities for integrated coherent ultra-violet sources and attosecond photonic devices. Up to now, most HHG studies have been limited to single crystals. Polycrystalline materials, which consist of many grains separated by grain boundaries and normally have random crystallographic orientations as well as scattering propagation trait, have rarely been explored for HHG although they have diversified properties. Understanding and predicting the HHG properties in polycrystalline nano-films rather than in pristine bulks are important owing to its merits of low cost and large deposition area. However, it is a formidable challenge when considering complex effects of crystallite size and texture, grain
boundaries, defects, surface states and nonlinear propagations. Here, we for the first time experimentally discover the correspondence between HHG in polycrystalline matters and macroscopic material parameters, to the best of our knowledge. Bright and long-term stable harmonics extending to 15th and 25th orders are demonstrated in polycrystalline cadmium telluride (CdTe) films with 6 nm and 50 nm thicknesses, respectively. It is found that the HHG strengths in the transmission and the reflection from the polycrystalline CdTe nano-films behave differently as a function of the material thickness in the range from 6 nm to 4 µm, which is highly correlated to the measured macroscopic conductivity. This work provides a simple gauge to study and predict HHG in complicated polycrystalline and amorphous nano-systems, and paves the way for novel strong-field nanophotonics based on polycrystalline nano-films.

High-harmonic generation (HHG) in solids is an emerging research frontier for applications in coherent spectral extension and rich physics investigations correlated to the electronics band structure, crystal orientation, and atomic interactions. HHG in solids has been investigated in bulk pristine crystals\textsuperscript{1-4}. On the other extreme, atomically thin two-dimensional materials have also served as a good platform to study the strong-field ultrafast dynamics including many-body interactions and induced distinctive symmetries\textsuperscript{5-7}. HHG from the intermediate thin films with thicknesses of a-few to few-hundred nanometers could not only enable the strong-field on-chip integrated photonics leveraging on semiconductor technologies\textsuperscript{8-10}, but also open up more advanced research possibilities such as attosecond control of intraband electron motion\textsuperscript{11}, ultrafast quasiparticle collision\textsuperscript{12}, direct measurement of Berry curvature in solids\textsuperscript{13}, and resonant enhancement by epsilon-near-zero effect\textsuperscript{14}. Moreover, HHG in solids is commonly explained with the origins of interband recombination induced by polarization between the conduction and the valence bands, and
intraband relaxation originated from non-parabolic band dispersion contributions. The microscopic mechanisms of HHG in solids have been investigated in pristine crystals and monolayer materials. HHG in polycrystalline and amorphous solids has also been demonstrated, however, the two microscopic HHG mechanisms are not applicable, since they are based on the Bloch theorem and electronic band structures of single crystals. It is a formidable challenge to understand or predict the HHG processes in polycrystalline solids when taking into account the complex effects of crystallite size and texture, grain boundaries, defects, surface states and nonlinear propagations, which hinders the development of strong-field research in polycrystalline nano-systems.

In this work, we report on experimental studies of bright HHG in polycrystalline cadmium telluride (CdTe) thin films, for the first time, in a broad thickness range over 6 nm to 4 µm. Pumped by a mid-infrared (MIR) femtosecond laser centered at 7.1 µm wavelength, the HHG in transmission (THHG) up to 25th and 15th orders are observed in CdTe ultra-thin films with 50 and 6 nm thickness, respectively, which bridges HHG in monolayer materials and thin films with hundreds-nanometer thickness. It is measured that the THHG exhibits the strongest HHG emission at two optimal thicknesses of 50 nm and 630 nm, while the intensity of HHG in reflection (RHHG) increases monotonically as the film thickness decreasing in the range of 4 µm to 30 nm. The thickness-dependent conductivity of the polycrystalline CdTe nano-films is acquired independently via two methods, namely four-point probe measurement and power transmittance measurement. Both THHG and RHHG strengths with the absorption deducted from the measurement are found to exhibit strong correlation with the measured macroscopic conductivity of the nano-films which are determined by microscopic factors such as the electronic motion of the bulk material, surface electronic structures and nonlinear propagation effect. This work opens up possibilities to investigate the microscopic
HHG processes through macroscopic material specifications in complicated condense matter systems. It also provides a new path towards the research of on-chip HHG and attosecond photonics based on CMOS-compatible polycrystalline CdTe thin films commonly used in the photovoltaic and X-ray detector industry, with the merits of low cost, large size, and good chemical durability.

In our experiment, CdTe nano-films are deposited on fused silica substrates by magnetron sputtering deposition technique which is commonly used in the semiconductor industry. The details of growth and characterization of polycrystalline CdTe thin films are presented in the Methods and Supplementary Information. CdTe is chosen for its extraordinary strong-field characters originated from a flat shape of band dispersion near the Fermi level and high carrier mobility, which has recently been observed in crystalline CdTe bulk solid. A MIR femtosecond laser centered at 7.1 µm with a pulse width of 180 fs at 50 kHz repetition rate is employed as the pump source, and both the THHG and RHHG are measured, as depicted in Figure 1(a). The photon energy of the pump laser is well below the direct bandgap of CdTe bulk crystal (around 1.44 eV) and thus the multi-photon ionization is negligible to explore the properties of HHG in CdTe nano-films. The XRD spectrum of 630-nm thick polycrystalline CdTe film is displayed in Figure 1(b). The main diffraction peak of <111> orientation at an angle of 23.75° dominates the XRD spectrum, while the zoom-in plot shows a serials of diffraction orientations, revealing the polycrystalline nature of the CdTe film. Figure 1(c) presents the THHG spectra of CdTe films with the thickness of 6 nm, 50 nm, and 630 nm. Up to 25\textsuperscript{th} (4.36 eV) harmonics are detected in the 630-nm-thick CdTe film at a peak driving intensity of 0.85 TW/cm\textsuperscript{2}. Remarkably, even the 6-nm-thick CdTe can support THHG up to the 15\textsuperscript{th} order with bright visible emission at a pump intensity of 0.4 TW/cm\textsuperscript{2}. This bridges the HHG from monolayer two-dimensional materials and thin films with hundreds nanometer thickness. Only odd harmonics are observed in our
measurements because of inversion symmetry in polycrystalline CdTe films, which is verified by the evenly distributed HHG strength along all the polarization directions of the driving field as the case in the atomic system (see Figure 2(c)). The scanning electron microscopy images of polycrystalline thin films with the thickness of 50 nm, 630 nm, 1.3 µm, and 4 µm are shown in Figure 1(d-g) manifesting randomly distributed grains which grow in size as the film thickness increasing. The distinct surface morphology and electronics states may influence the HHG strength, especially for THHG while the fundamental and harmonics pulses are propagating in polycrystalline films. The photos of the bright visible THHG emission from the CdTe films with the thickness of 6 nm, 50 nm and 630 nm are captured and shown in Figure 1(i-k). It is worth noting that comparable HHG emission from CdTe films with 50 nm and 630 nm thickness is observed from the photos, because the 630-nm-thick polycrystalline CdTe film has much bigger grain size and rougher surface morphology, so that stronger HHG emission is scattered to the CCD which is located at a small noncolinear angle with the pump laser. The fused silica substrate is also excited under the same experimental conditions as a comparison, and it exhibits neither visible emission nor infrared spectral signal, as shown in Figure 1(h). We suppose that the extraordinary HHG emission from the polycrystalline CdTe nano-films is attributed to both the excellent electronic characteristics inherited from the crystalline bulk which are flat shape of band dispersion near the Fermi level (The calculations of atomic and electronic structures of bulk CdTe crystal could be found in Supplementary Information Section 1) and high carrier mobility (The carrier mobility of CdTe pristine crystal is 1100 cm²/(V·s)²¹ which is 2-orders higher than that of MoS²⁹), and the unique features including surface electronic structures, microscopic light propagation, and electronic motion in the polycrystalline nano-films. More studies are therefore conducted to explore the physical mechanisms behind.
The individual harmonics strengths of 9\textsuperscript{th}-17\textsuperscript{th} orders from the polycrystalline CdTe film with a thickness of 50 nm are plotted on the logarithmic scale versus the driving-laser intensity ranging from 0.3 to 1.2 TW/cm\textsuperscript{2}, as shown in Figure 2(a). The power-law fit curves reveal values of exponent $n \sim 2.5$ except for the 9\textsuperscript{th} order with $n \sim 3.5$ (the fluorescence signal), which clearly demonstrates the non-perturbative behavior of HHG in CdTe films. As shown in Figure 2(a), the 50-nm-thick CdTe film can support harmonics up to the 13\textsuperscript{th} order at a pump intensity of 0.3 TW/cm\textsuperscript{2}. At a higher pump intensity, the long-term stable HHG emission is observed from the polycrystalline CdTe nano-films as presented in Figure 2(b). The 50-nm-thick CdTe film is excited at a MIR laser intensity of 1 TW/cm\textsuperscript{2}, and harmonics signals of 9\textsuperscript{th} to 17\textsuperscript{th} orders are recorded for 10 minutes. Remarkably, stable long-term HHG output almost without decline is measured, which implies that there is no damage for the polycrystalline nano-film exposed in such a strong field.

To investigate the polarization of HHG from polycrystalline CdTe films and the dependence on the driving laser polarization, the harmonics intensity of 9\textsuperscript{th}-17\textsuperscript{th} orders from a 50-nm-thick CdTe film are measured by fixing the pump polarization and rotating the CdTe nano-film about the normal. Figure 2(c) shows that the THHG from the polycrystalline CdTe nano-film is insensitive to the polarization of the driving laser filed. Furthermore, as presented in Figure 2(d), the polarization of the harmonics from polycrystalline CdTe films follows the polarization of the driving laser, which indicates the atomic inversion symmetry in polycrystalline CdTe thin films.

The harmonic strengths with respect to the film thickness are investigated to understand the complex dynamics of HHG in polycrystalline nano-films. Figures 2(e, f) are the measured spectra plotted on linear scale of THHG and RHHG from CdTe films with a broad range of thickness ranging from 30 nm to 4 µm. The THHG is detected directly behind CdTe films at a pump intensity of 1 TW/cm\textsuperscript{2}. As presented in Figure
2(e), THHG of 11th-15th orders in CdTe thin films has the maximum strength at a thickness of 50 nm, and exhibits an oscillation with respect to the film thickness. The RHHG is also measured 10-mm in front of the CdTe nano-films with angles of incident and reflection both as ~ 45°, under a pump intensity of 0.57 TW/cm². As plotted in Figure 2(f), unlike the THHG, the RHHG shows a broadband fluorescence peak at wavelengths ranging from 400 to 800 nm, which is attributed to surface defects states from polycrystalline thin films. Intriguing, the RHHG of CdTe nano-films has the maximum at a thickness of 30 nm and decreases monotonically as the thickness increasing. It is important to note that absorption should be accounted for the THHG with harmonics orders above the bandgap. We observe that the band edge of polycrystalline CdTe nano-films moves to shorter wavelengths as the film thickness getting thinner (see the measured transmission spectra of polycrystalline CdTe films with various thicknesses in Supplementary Information Figure 4), which is attributed to the quantum size effect. This agrees with the observed blue shift of the fluorescence envelopes as the thickness decreases, shown in Figure 2(f). Therefore, distinct absorption of individual CdTe films with different thicknesses should be deducted to reveal the true THHG strength.

The true strengths of THHG and RHHG in polycrystalline CdTe films without absorption are therefore properly measured and calculated as presented in Figure 3, to analyze the thickness dependence of HHG from CdTe nano-films. The harmonics ranging from 11th to 17th orders are measured and compared with broad film thicknesses over 30 nm to 4 µm. We calculate the absorption coefficients of CdTe nano-films with different thicknesses based on the measured transmission curves, and then deduce the true THHG strengths without absorption based on the measured THHG intensity, as shown in Figure 3(a-d) (the details of the calculation are presented in Supplementary Information Section 4 and 7). THHG shows a clear trend of oscillation with various film thicknesses, with two maximums at 50 nm and 630 nm. To understand the origin of oscillation,
it is noted that HHG strength is directly linked to the microscopic electric current density and thus the conductivity in the medium by \( \text{HHG}(\omega) = \left| \text{F.T.} \left( \frac{\partial}{\partial t} \int j(r, t) d^3r \right) \right|^2 \), where \( j(r, t) \) represents the electric current density and F.T. denotes the Fourier transform. The current density and conductivity are then linked by \( j(r, t) = \sigma E(r, t) \), where \( \sigma \) and \( E(r, t) \) are the conductivity and electric field, respectively. We therefore suggest that in CdTe films with the thickness of few nanometers the HHG strength increases with the film thickness, and it soon saturates at around 50 nm and starts to decline, due to the reduced conductivity and current density in the CdTe film as the thickness increasing. This agrees well with the recent theoretic prediction\(^{20}\). While the film thickness is further increasing beyond around 500 nm, we believe enlarged grain boundaries and defects states of the polycrystalline thin films start to play a more dominating role to enhance the current density so as the harmonics strengths while the fundamental and harmonic pulses propagating in the CdTe films. Thus a second peak of THHG is exhibited. Notably, HHG enhancement by defects in solids has been reported before\(^{26-28}\). It is worth mentioning that in pristine crystals, it has been experimentally observed that the THHG strength from a GaAs crystal drops as the thickness increasing from 45 µm to 650 µm, which is associated with the nonlinear propagation in the periodic solids\(^{29}\). It is thus suggested that the randomly distributed grains and voids in the polycrystalline CdTe film modify the dynamics of nonlinear propagation effect which impact on the harmonics generation. On the other hand, unlike THHG, the RHHG signal declines as the thickness increasing from 30 nm to a few micrometers as presented in Figure 3(e-h). We suggest that this is because RHHG is less influenced by the nonlinear propagation effect imposed by the polycrystalline grain and voids, thus a monotonic decline of HHG strength is observed, following the trend of the reduced conductivity and current density in the CdTe films. Notably, RHHG from extremely thin CdTe films (few-nanometer) is not measured due to the difficulty in collecting the weak RHHG signal in the pump
incidence direction.

In a macroscopic perspective, the distinct trends of harmonics in polycrystalline CdTe nano-films may be related to the conductivity and current density of thin films with different thicknesses. To explore the link between the trend of HHG and the macroscopic parameters with respect to the polycrystalline film thickness, we thus measure the conductivity of CdTe films with various thicknesses using two methods independently. The four-point probe method is employed as the direct measurement of the conductivity, while the power transmittance measurement is conducted as a replication proof according to $A_L^{(l)}(\omega) = \left(1 + \frac{2\sigma(\omega)}{c} \right)^{-1} A^{(l)}(\omega)$, where $A^{(l)}$ and $A_L^{(l)}$ denote the linear components of the incident and transmitted fields, $d$ represents the thickness of the thin film, and $c$ is the speed of light. Two wavelengths of 1 µm and 2.4 µm are used in the power transmittance and reflectance measurement for their negligible absorption in the fused silica substrate. As shown in Figure 4, the two methods demonstrate generally the same trends with harmonics strengths versus the thickness of polycrystalline CdTe films. In particular, the conductivity scales inversely proportional to the thickness as shown in Figure 4(b, e, h), which is consistent with the trend of RHHG in Figure 3(e-h). This also verifies our assumption that the RHHG is mainly generated in the front few nanometers, and not influenced a lot by the nonlinear propagation effect. On the other hand, the product of conductivity and the film thickness, especially measured by the power transmittance method as presented in Figure 4(f, i) displays qualitatively similar oscillation trend with that of the THHG and shows two local maximums at 50 nm and 600 nm. Notable, the polycrystalline CdTe thin films with the thickness of 50 nm and 600 nm exhibit two peaks of the measured reflected power at both wavelengths of 1 µm and 2.4 µm, as shown in Figure 4(d, g), which implies the polycrystalline CdTe thin films at the particular thicknesses have significantly higher conductivity, reaching the level of semimetals. These results suggest that the conductivity
of the film which is not only determined by the electronics structures, but highly influenced by the surface morphology and defects states in a material is a key factor of HHG in polycrystalline films. It could serve as a simple gauge in a macroscopic view to analyze the HHG performance in complicated material systems.

In conclusion, we experimentally observe high harmonic generation in both transmission and reflection directions from polycrystalline CdTe films with various thicknesses ranging over 6 nm to 4 µm, pumped by a MIR femtosecond laser at 7.1 µm center wavelength. The non-perturbative HHG from 6-nm and 50-nm thick CdTe films extends to 15th and 25th order-harmonics, respectively. It is found that THHG and RHHG behave distinctly with respect to the thin film thickness. THHG is peaked at two thicknesses of 50 nm and 630 nm, while RHHG decreases monotonically as the thin film thickness increases. The conductivity of the nano-films is measured to reveal the origin of variation in HHG strengths with respect to the nano-film thickness, which suggests that the HHG from the polycrystalline CdTe nano-films is closely related to the macroscopic conductivity of the thin films. This work demonstrates exciting possibilities in HHG and attosecond nanophotonics in the polycrystalline thin films, such as CdTe as demonstrated in this work, with extraordinary strong-field characteristics and stable chemical properties, as well as mature fabrication technologies. Moreover, this work provides a possibility of analyzing and predicting the HHG performance of complicated material systems based on macroscopic parameters, which is otherwise difficult to realize by the existing microscopic models.

Methods

Film growth and characterizations. The CdTe films are fabricated by magnetron sputtering deposition technique at a temperature of 235 °C on 0.5-mm-thick fused silica (JGS2) substrates. The thickness of the
CdTe films is controlled by the sputtering time and measured by atomic microscopy (AFM) (More information could be found in Supplementary Information Section 3). The surface morphology of the polycrystalline CdTe films is characterized by a scanning electron microscope (SEM) (More information could be found in Supplementary Information Section 2). The crystal orientation of CdTe films is measured by X-ray diffraction spectroscopy (XRD) using Cu-Kα radiation with 2θ of 10–90°. The transmittance of CdTe films with different thickness is characterized by UV-Vis-NIR spectrometry with a wavelength range over 250 nm to 2500 nm (See Supplementary Information Figure 4).

**High-harmonic generation pumped by the MIR femtosecond laser.** The pump source of HHG in CdTe is a MIR optical parametric amplifier (OPA) driven by a commercial Yb-doped regenerative amplifier (Pharos) with a maximum 20 W power. The MIR OPA consists of two-stage amplifiers based on LiGaS₂ crystals, as shown in the experimental schematic in Supplementary Figure 5. The MIR power up to 280 mW at 7.1 μm could be produced. The temporal duration of the MIR pulse is measured by a home-built interferometric autocorrelator (IAC) and reconstructed through a genetic algorithm based on so-called “evolutionary phase retrieval from IAC (EPRIAC)” algorithm. 180 fs pulse width is estimated with more details shown in Supplementary Figure 6 (b, c). An infrared camera (Dataray, WinCamD-IR-BB-7.5 system) is employed to check the MIR beam profile and measure the beam size as shown in Supplementary Figure 6 (d).

CdTe thin films with various thicknesses ranging from 6 nm to 4 μm grown on fused silica substrates are pumped at an intensity ranging from 0.3 TW/cm² to 1.2 TW/cm² focused by zinc selenide lenses with a focal length of 100 mm. The HHG spectra are measured by two different Ocean Optics spectrometers (USB 2000+ and Maya 2000 Pro). The spectra are recorded by directly coupling the emission into a silica fiber with a 400-μm core diameter at the back surface of the CdTe crystal for the MIR and visible-to-ultraviolet harmonics.
measurement, respectively. The long-term stability of generated harmonics signals from the CdTe films with representative thicknesses of 50 nm, 630 nm and 2.32 µm is measured for 10 minutes by recording the harmonics spectra once per 20 seconds.

**Data availability.** The data that support the findings of this study are available from the corresponding author upon request.

**Reference**

Acknowledgement:
This work was supported by National Natural Science Foundation of China (62075144, 12175157), Sichuan Outstanding Youth Science and Technology Talents (2022JDJQ0031), and Engineering Featured Team Fund of Sichuan University (2020SCUNG105).

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Author contributions:
H. K. Liang conceived and designed the experiment. Z. Long and H. Yang carried out the experiment of HHG measurement. Z. Long, H. Yang, Sen Lin, Dewei Zhao and Lili Wu grew the CdTe thin films. Zi-Yu Chen conducted the theoretical simulation. K. Tian and L. He built the MIR OPA. H. K. Liang, Z. Long, H. Yang, Zi-Yu Chen and Han Wu wrote the manuscript. Z. Long and H. Yang contribute equally. All authors discussed the results and contributed to the manuscript.
Figures

Figure 1 | The experimental schematics, X-ray diffraction (XRD) spectroscopy, and measured high-harmonic spectra and corresponding emission photos for HHG in polycrystalline CdTe nano-films. a, Simplified schematic diagram of the HHG measurement for both THHG and RHHG. b, The measured XRD spectrum of the 630-nm thick polycrystalline CdTe film showing a dominating crystal orientation of <111>. The zoom-in plot shows other crystal orientations in polycrystalline CdTe film. c, Measured spectra of HHG in transmission from polycrystalline CdTe films with the thickness of 6 nm, 50 nm and 630 nm, pumped at 7.1 µm wavelength. Harmonics up to 15th and 25th orders are observed from the 6-nm, 50-nm and 630-nm thick CdTe films, with a pump intensity of 0.4 TW/cm², 0.85 TW/cm² and 0.85 TW/cm², respectively. The 50 nm thick polycrystalline CdTe film has the strongest HHG strength. d-g, Scanning electron microscope (SEM) images of polycrystalline CdTe films of representative thickness of 50 nm, 630 nm, 1.3 µm and 4 µm. The scale bars represent the lengths of 1 µm. SEM images of CdTe films thinner than 30 nm are not included.
as the polycrystalline CdTe grains could not be differentiated clearly from the gold particles from the metalized coating while taking the
SEM images. The photo of the fused silica substrate pumped at 7.1 µm with an intensity of 1.3 TW/cm². No visible emission or
infrared spectral signal is observed from the substrate. The photos of the HHG emission from polycrystalline CdTe thin films with a
thickness of i, 6 nm, j, 50 nm and k, 630 nm, pumped at 7.1 µm with an intensity of 0.3 TW/cm², 1.3 TW/cm², and 1.3 TW/cm²,
respectively. The photos show comparable HHG emission from CdTe films with 50 nm and 630 nm thickness because 630 nm thick
polycrystalline CdTe film has much bigger grain size and more rough surface morphology (as shown in Supplementary Information
Section 2), so that stronger HHG emission is scattered to the CCD which is located at a small noncolinear angle with the pump laser.
Figure 2| Dependence of HHG on pump intensity, long-term measurement of HHG, polarization characterization of HHG and spectra of HHG in transmission (THHG) and reflection (RHHG) in CdTe nano-films with a few representative thicknesses. a, Logarithmic plot of the measured harmonics signals with 9th to 17th orders with respect to the excitation intensity in polycrystalline CdTe
films with a thickness of 50 nm. Fitting of the experimental data based on the power-law equation \( I \propto I_0^n \) yields the corresponding exponents (\( n \)) for each harmonic (solid lines) ranging from 2.1 to 3.5, which deviates strongly from the conventional perturbative nonlinear response optics \( n = k \) (dashed lines, \( k \) is the harmonic order). b, The long-term measurement of the THHG intensity from the CdTe thin film with a thickness of 50 nm. The harmonics of 9th to 17th orders are recorded for 10 minutes with stable strengths. c, The harmonics strengths of 9th-17th orders with respect to the polarization of pump field varying from 0° to 180°. d, The THHG spectra in polarizations parallel and perpendicular to the pump field. The harmonics with vertical and horizontal polarizations are separated by using a FLP20-VIS polarizer, and the pump intensity is 0.35 TW/cm². Spectra on linear scale of e, THHG and f, RHHG, with the thicknesses in the range of 30 nm to 4 µm. The THHG and RHHG spectra contain 11th to 15th and 9th to 17th orders harmonics, respectively. THHG strength is peaked at 50 nm and oscillates with respect to the film thickness. While the RHHG has the maximum at a thickness of 30 nm and decreases monotonically as the thickness increasing. Clear fluorescence envelopes spanning from 400 nm to 800 nm can be seen from RHHG spectra, which is attributed to the surface defect states in the polycrystalline nano-films. There exhibits blue shift of the fluorescence envelope as the thickness decreases. This is because the band edge of CdTe polycrystalline films move to shorter wavelength when the film thickness getting thinner (see the measured transmittance of various thicknesses in Supplementary Information Figure 4)
Figure 3] Comparison of the THHG and RHHG strengths without absorption from polycrystalline CdTe films as a function of thickness. **a-d**, THHG of 11$^{th}$ to 17$^{th}$ orders in CdTe films with thickness ranging from 30 nm to 4 µm. There are two maximums located at 50 nm and 630 nm. **e-h**, The same as **a-d** but showing RHHG strengths. The harmonics strength of RHHG gradually decreases as the thickness getting thicker. The absorption coefficients of CdTe nano-films with different thicknesses based on the measured transmission curves are calculated, and then the THHG strengths without absorption is obtained. (The details of the calculation could be found in Supplementary Information Section 4 and 7.) The dashed curves with arrows are guide for the eye showing the trends of THHG and RHHG varying as the thickness of CdTe films increasing.
Figure 4: Comparison of measured conductivity and the product of conductivity and the film thickness as a function of the polycrystalline CdTe thickness. 

- **Figure 4a**: Schematic diagram of the conductivity measurement via four-point probe method. The measured conductivity  
- **Figure 4b**: and the product of conductivity and the film thickness  
- **Figure 4c**: via four-point probe method. Measured power transmittance and reflectance of  
- **Figure 4d**: 1 µm and  
- **Figure 4g**: 2.4 µm wavelengths as a function of the CdTe film thickness, and corresponding calculated conductivity  
- **Figure 4e**: and product of conductivity and the film thickness  
- **Figure 4h**: and  
- **Figure 4i**: The dashed curves with arrows are guide for the eye showing the trends of macroscopic conductivity varying as the thickness of CdTe films increasing, which qualitatively agrees with the measured trends of RHHG and THHG as shown in Figure 3.
Supplementary Files

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