Terahertz-assisted even harmonics generation in silicon

Highlights
- THz-assisted even harmonics generation in Si is observed and characterized.
- Ultrafast symmetry control of centrosymmetric crystal Si is demonstrated.
- Effect of THz and Si orientation on even harmonics generation is revealed.

Ding et al., iScience 25, 103750
February 18, 2022 © 2022 The Author(s).
https://doi.org/10.1016/j.isci.2022.103750
iScience

Article

Terahertz-assisted even harmonics generation in silicon

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SUMMARY

When a biased electric/light field is applied to centrosymmetric crystals like silicon, the broken symmetry creates even-order harmonics radiation which can reveal key insights into the material. Recently, the second harmonic has been generated by THz-induced symmetry breaking, but the observation of higher-order radiation remains largely unexplored. Here, we demonstrate picosecond-level ultrafast, nondestructive symmetry manipulation of silicon crystal by using a 500 kV/cm intense terahertz (THz) electric field. The THz-induced fourth harmonic of the infrared probe is also observed and characterized for the first time. In addition, we find that the even-order harmonics show no dependence on the THz field direction thus it allows for sub-cycle symmetry manipulations. Our study paves the way toward ultrafast all-optical crystal symmetry control in the future high-speed electronics and photonics.

INTRODUCTION

Symmetry is a fundamental property of crystals that can leverage dynamical functions in photonic applications (Armstrong et al., 1962; Leuthold et al., 2010). Especially in the nonlinear optics regime, manipulation of crystal symmetry can provide unique ways for modifying the crystal performances in structural phase transition (Collet et al., 2003), second harmonic generation from centrosymmetric materials (Timurdogan et al., 2017), and enhancement of nonlinear properties such as frequency conversion (Vicario et al., 2014), pulse stretching (Kobayashi et al., 2016), electro-optic modulation (Chen et al., 2001), and self-detection of optical beam (Siviloglou et al., 2006). The earliest symmetry breaking report can be traced back to the 1990s, when Ivanov and Corkum first demonstrated harmonic control by using a strong laser pulse experimentally (Ivanov and Corkum, 1993). Afterwards, tremendous interests have been aroused in the searching for symmetry control means and associated new phenomena never have been charted before (Mueller et al., 2015), for example, the discovery of exceptional points in plasmonics by spatial symmetry breaking (Park et al., 2020). And the recent observation of giant anisotropic terahertz (THz) nonlinear currents with vanishing scattering are also disclosed by breaking inversion symmetry in a centrosymmetric Dirac material ZrTe5 (Luo et al., 2021).

So far, laser pulses and DC current have been the most widely used means for breaking symmetry (Cazzanelli and Schilling, 2016; Shamim et al., 2014). However, in the most advanced electronics and photonics in particular, both these two above approaches fall short either in the modulation timescales or in terms of no destruction. The frequency of electric dipole associated with the charge carriers moving in the ensuing two half-cycles of the externally applied field prescribes the speed limit of the symmetry breaking time, which corresponds exactly to a special band in the light spectrum—the Terahertz band (0.1–10 THz, or 1011–1012 Hz) (Salén et al., 2019). Meanwhile, as a relative new driving source, THz has many unique advantages (Kampfrath et al., 2013), such as photon energy that corresponds to vast low energy modes in materials like spin, carrier moving, phonon oscillations, and shall not cause thermal deposition damage to materials. Recently, with the THz electric field achieving higher strength (exceeding 1MV/cm [Fülöp et al., 2019]), major progress has been made in the THz field control of matter (Salén et al., 2019) including spin flipping (Schlauzerer et al., 2019), phonon mode excitation (Afanasiev et al., 2021), and carriers control in semiconductors (Fausti et al., 2011; Matsunaga et al., 2014; Mitrano et al., 2016).

In symmetry control, THz-field-induced second harmonic generation has also been observed from non-centrosymmetric ferroelectric (Grishunin et al., 2017) and antiferromagnetic materials (Ovchinnikov et al., 2016). These works lay the firm grounds for pursuing ferro-/magneto-based ultrafast devices that may

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https://doi.org/10.1016/j.isci.2022.103750
work at picosecond processing speed. However, for the most commonly used material that underpins most photonic components (Bogaerts and Chrostowski, 2018) and the modern semiconductor industry (Kim et al., 2020; Yang et al., 2020), a lot more possibilities could be expected if the symmetry of the centrosymmetric Si crystal can be modulated in a nondestructive and ultrafast manner. For this purpose, electric-field-assisted even-order harmonics generation is reported in Si and ZnO crystal (Vampa et al., 2018). In addition, a static electric field is used to induce $c^{(2)}$ in a Si waveguides and trigger the second harmonic generation (Timurdogan et al., 2017). Later on, up to 15MV/cm extremely intense THz electric field has been used to study the saturation of the second harmonic in a 245-μm-thick p-doped Si wafer (Ovchinnikov et al., 2019). Nevertheless, the picosecond-level ultrafast symmetry manipulation and the observation of higher even-order harmonics have not been demonstrated yet.

In this paper, we demonstrate experimentally the picosecond ultrafast symmetry manipulation accompanied with the second and fourth harmonics generation by applying an intense THz electric field to a Si crystal film. The transient generation of even-order harmonics along THz electric field is measured. For Si, a typical central symmetric crystal without even-order polarization, the intensity of the generated second and fourth harmonics are affected by the incident THz electric field polarization which determined the even-order nonlinear polarity. It is believed that this transient all-optical crystal symmetry control technology will play a significant role in high harmonics generation, optical frequency conversion, THz electric field measurement, and other ultrafast electro-optical applications.

**RESULTS AND DISCUSSION**

**Experiment setup**

In our experiments, a thin (2 μm-thick, 100-cut), free-standing, crystalline silicon film is exposed to intense THz fields generated by optical rectification in an organic crystal DSTMS (4-N, N-dimethylamino-4′-N′-methyl-stilbazolium 2,4,6-trimethylbenzenesulfonate). As illustrated in Figure 1, the pump-probe method is employed for the detection of the even-order harmonics generation which is most susceptible to the symmetry changes. For the THz pump, Figure 1A and 1B, respectively, show the time-dependent waveform,
and the corresponding spectrum measured by replacing the Si sample with a 100-μm-thick electro-optic crystal GaP. When focused normally into a spot diameter of 600 μm, the single-cycle, 3-THz-centered THz pulse peak would result in a peak field amplitude of 500 kV/cm on the Si crystal. However, for the weak copropagating 120-femtosecond-duration, 1450-nm wavelength-centered infrared probe, the focus spot has a diameter of 76 μm, thus allowing for a homogeneous region that is detected by the probe beam.

During the experiment, both the THz pump and the infrared probe beams are set to linear polarization parallel to the [100] (y axis, defined in the laboratory frame) of the Si crystal. Meanwhile, a filter is also used to block the infrared pulse behind the DSTMS. The intensity of the THz and the infrared beams on the silicon are 0.66 GW/cm² and 61 GW/cm², respectively. By changing the delay between the pump and the probe, the time-dependent evolution of the harmonic generation can be recorded by the optical spectrometer (USB4000, Ocean Optics Inc.) placed behind the Si crystal. When there is no THz presented or if the two beams are not overlapped in time, only odd-order harmonics are detected, attesting to the inversion symmetry of the silicon crystal.

Even-order harmonics generation

Phenomenologically, in centrosymmetric crystals like Si, inversion symmetry can be lifted by applying a biased electric field, which give rise to high-order expansions of the polarization $P(\omega)$. For the even-order nonlinearity, it depends mainly on the additional THz electric field and the odd-order susceptibility $\chi^{(n)}$. Specifically, by assuming a driving laser field of $E(\omega)$, the THz-induced even-order polarization can then be expressed as a power series in the $E(\omega)$ as:

$$\sum_{n=1}^{\infty} P(2n\omega) = \sum_{i=1}^{\infty} \chi^{(2i+1)}(\omega) E(\omega)^{2i+1} E_{\text{THz}}$$

(Equation 1)

where $\chi^{(2i+1)}$ is the $(2i+1)$ order susceptibility, $E_{\text{THz}}$ is the instantaneous THz electric field, and $E(\omega)$ refers to the electric field of the probe beam. Here, by using an intense THz pulse, both the emergence of second

![Figure 2. THz-induced even-order harmonics generation in Si crystal](image_url)
and fourth harmonics generation are detected in the Si crystal, which originated from the even-order polarization that are specified by Equation 1.

Specifically, for the Si crystal with cubic symmetry, the contribution to second-order harmonic generation in our experiment geometry comes from three particular components - $\chi^{(3)}_{xycz}$, $\chi^{(3)}_{xxyz}$, and $\chi^{(3)}_{xyzx}$ of the fourth-rank nonlinear susceptibility $\chi^{(3)}$. Besides, considering that the THz beam and the IR are collinear and both incident normally to the Si sample along the x axis, $\chi^{(3)}_{xxxx}$ is an independent value regardless of the configuration of the polarization angles between the THz and IR or the rotation of Si sample. Therefore, for the second harmonic, Equation 1 can be further written in the following format:

$$E_{x}^{2\nu} \propto \left[ \left( \chi^{(3)}_{xxyx} + \chi^{(3)}_{xxyz} \right) \cos^{2} \varphi + \left( \chi^{(3)}_{xxyz} + \chi^{(3)}_{xyzx} \right) \cos^{2} \theta + \chi^{(3)}_{xxxx} \right] E_{x}^{\nu} E_{x}^{\text{THz}}$$

(Equation 2)

where $\varphi$ denotes the polarization angle between THz and IR beams, and $\theta$ is the rotation angle of the Si sample (Here, 0 represents the [100] being parallel the y axis). The expression for the fourth harmonic can be given in a similar way. Based on Equation 2, the even-order harmonics is the strongest when the THz has the same polarization with the IR beam.

Figure 2A shows a snapshot spectrum of the generated second and fourth harmonics when the THz polarization is synchronized with the infrared and is parallel to the y axis. No even-order harmonics are found when there is no THz pulse on the Si sample. Because the probe light is 1450-nm wavelength centered, the second and fourth harmonics are found respectively around 745 and 365 nm with the amplitude of second harmonic about 16 times greater than that of the fourth harmonic. It should be noted that, for our 2-μm-thick Si sample, the optical transmission of the second harmonic of our probe beam is approximately 76.6%, whereas the transmission decreases to vanishingly small ($2.724 \times 10^{-19}$) for the fourth harmonic (Schinke et al., 2015). In other words, the fourth harmonic we detected arises almost entirely from the tens-nanometer depth of rear surface of the Si sample, showing a major difference from that of the second harmonic generation.

This distinction of signal range can not only lead to variations in the intensities of second and fourth order harmonics but also the phase-matching conditions and spectral shape of the second and the fourth harmonic signal. Specifically, for the phase-matching conditions, coherence length of the second harmonic signal is estimated by $L_{c} = 2/[2k_{1} - k_{2}]$ to be 1.792 μm, which is close to the thickness of our Si sample, thus obviating the destructive interference of the generated second harmonic. However, for the fourth harmonic signal, dephasing would be automatically eliminated inside the tens-nanometer depth of the Si rear surface. On the other hand, the signal range difference could also contribute to the spectral variance of the second and the fourth harmonics, as discernible in Figures 2B and 2C. This spectral broadening is attributed to two origins: firstly, the IR probe beam itself is not Fourier-transform-limited; secondly, for the fourth harmonic, the probe beam would accumulate an extra spectral broadening in front of the sample before it arrives at the Si rear surface, thus gaining extra spectral chirping than the second harmonic signal.
The time evolution of the second and fourth harmonics generation is recorded by scanning the delay between the THz pump and the infrared probe. This is depicted in Figures 2B and 2C where the harmonics signals are compared in align with THz waveform (Figure 2D) characterized by the electro-optical sampling (Wu and Zhang, 1995). From these results, it is found that the harmonics display an instantaneous imitation of the THz envelop with their peaks appearing almost exactly at the peaks or troughs of the THz waveform. The repeatability of this phenomenon verifies that the THz-induced symmetry breaking is a nondestructive process in our experiment. In addition, the intensity of the even-order harmonics varies with the strength of the THz electric field.

To further extract the reliance of even-order harmonics intensity on the THz electric field, second harmonic signal is plotted as a function of the THz electric field in Figure 3. From the fitting result, the second harmonic intensity shows a quadratic dependence on the driving THz electric field. This is in consistence with Equations 1 and 2 because the harmonics signal intensity is proportional to the square of the field-induced even-order polarization. No saturation of the second harmonic is found under our relatively moderate THz electric field (peak intensity of 500 kV/cm). However, for the fourth harmonic which is expected to trace the quartic power of the THz electric field, the signal-to-noise ratio (SNR) of the fourth harmonic (6:1, as in comparison to the 120:1 of the second harmonic) has limited our verification. This verification of higher-order harmonics could be fulfilled with increased driving THz intensity and laser system repetition rate to optimize the SNR in the feature measurements.

Polarization dependence on the probe beam

Microscopically, the even-order harmonics generation arises from the asymmetric electron motions in the two neighboring superimposed light half-cycles, regardless of whether it is an interband or intraband process. In this work, to further explore this asymmetry dependence on the angle between the THz pump and the infrared probe, the polarization of probe beam is rotated by 90° (along the z axis) with the help of a half-wave plate, whereas the polarization of the THz electric field is maintained along the y axis.

Figure 4. Comparison of the second (left) and fourth harmonics (right) while the infrared probe light is rotated from parallel to perpendicular

(A and D) show the second and fourth harmonics evolution when the IR beam polarization is perpendicular to THz beam polarization (denoted as ⊥).
(B and E) show the second and fourth harmonics when the IR beam polarization is parallel to THz beam polarization (denoted as ||).
(C) Depicts the corresponding THz waveform.

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By setting this perpendicular polarization, the even-order harmonics spectrum can reveal a glimpse of the atomic orbital symmetry perturbations inside the silicon crystal that are resultant from the quadrature components of the THz field (Niikura et al., 2010).

In Figure 4, we show in columns the second and fourth harmonics evolution with the probe perpendicular (denoted as $\perp$) or parallel (denoted as $\parallel$) to the THz pump. From these results, despite the second and fourth harmonics are detected in both situations, their intensities in the perpendicular polarization exhibit a sharp decrease (approximately 10 times) as in comparison to that of the parallel polarization. This contrast can be ascribed to the phase difference $\sigma$ accumulated by the electrons that undergo the two neighboring superimposed light half-cycles. In particular for the THz pump, the long wavelength/period of the THz wave allows the infrared probe to imprint a much more profound asymmetry in the electron orbital. In addition, according to the Equation 2, when $\theta = 0$ (the Si sample is fixed as [100] parallel to the y axis), and $\varphi = 0$ (I$\parallel$ THz), the generated even-order harmonics intensity becomes the strongest, whereas it turns into the weakest when $\varphi = 90$ (I$\perp$ THz), showing consistence with our experimental findings.

**Reliance on the Si crystal orientation**

The reliance of the even-harmonics intensity on the Si crystal orientation is also investigated while fixing the polarization of THz and the infrared to be parallel to the y axis. As shown in Figure 1, 0° is defined as the $<100>$ direction of Si crystal, and the Si sample is rotated clockwise from 0° to 180° between which the second and fourth harmonics intensities are recorded in time by every 30° increment.

**Figure 5. Dependence of even-order harmonics generation on the Si crystal orientation**

In this case, the THz pump and the infrared probe are polarized along $<100>$ direction of Si (0°).

(A) shows the second harmonic variations as the Si changes angles while (B) shows that of the fourth harmonic. (C) depicts the corresponding THz waveform.
Figure 5 shows the time-dependent intensity changes of the second and fourth harmonics as a function of the different Si sample angles. From these measurement results, several main features of the spectrum are clear: firstly, the waveform synchronicity of the even-order harmonics with the THz waveform is preserved throughout all Si angles. Secondly, the intensity of the second and fourth harmonic both decreased as the Si sample is rotated from 0° to 90°, and then returned to approximate its original value as the angle increased to 180°. In the process of rotating the Si angle, the polarization coefficient of THz electric field direction is changed, and the intensity of even-order harmonics caused by the incident THz electric field is naturally changed. Moreover, another essential feature of the even-order harmonics concerns with the peak positions disregarding the THz filed direction. That is, the even-order harmonics intensities always appear at the peaks or the troughs of the THz field. To take a step further, this phenomenon also presents one major difference between odd- and even-order harmonics. As shown in Figure 6A, for the odd-order harmonics, owing to face-centered diamond-cubic structure of the Si crystal, a quadruple symmetry is manifested while rotating the sample by one circle. Or in other words, the odd-order harmonics repeat itself at every 90°, just as analogous to the structure of the silicon structure. However, this quadruple symmetry turns into 2-fold for the even-order harmonics (Figure 5B) that are induced by the transient-intense THz electric field, which lacks the quadruple symmetry. This symmetry change can be phenomenologically interpreted by the bidirectional displacement of the silicon structure under the impact of the linear polarized THz field. Because the symmetry characteristic of the harmonic signals depends on the transient crystal structure, the THz field imposes a linear polarization of the silicon exactly alongside its electric fired direction, thus turning the quadruple symmetry into 2-fold. On the other hand, this symmetry change can also find explanations in Equation 2: When ϕ=θ (IR||THz), as the Si sample (θ) rotates from 0° to 360°, the generated even-order harmonics acquire the symmetry of COS2θ which is exactly 2-fold. Hence, while the even-order harmonics polarization relies solely on the amplitude of the THz electric field, the symmetry disparity between the odd- and even-order harmonics also verifies the symmetry control that are induced by the THz field.

Conclusions
In conclusion, we have demonstrated picosecond-level ultrafast, nondestructive symmetry manipulation of Si crystal. The second and fourth harmonics are observed and characterized in terms of their dependence respectively on the light field polarizations and the crystal orientations. For the centrosymmetric Si crystal studied in this work, it is found that the even-order harmonics have the maximal intensity when the THz polarization is parallel to that of the infrared probe and is aligned along the <100> direction of the Si crystal. Furthermore, by scanning the time evolution of the even-order harmonics, their intensities exhibit an instant imitation of the amplitude THz field strength rather than the energy. And no reliance on the direction of the THz electric field is found in the generation process of the even-order harmonics. This feature unequivocally indicates the possibility to a symmetry manipulation at sub-cycles within the THz wave. Meanwhile, the generation of higher-order harmonics has far more profound implications in the exploration of ultrafast dynamics inside a light-field-driven solid as well as their relative applications in the integration of electronics and optics. And this will provide a new idea for the...
generation of UV, including extreme UV light sources, and theoretical support for the development of semiconductor combining with corresponding silicon photonic technology. The ultrafast, nondestructive symmetry control demonstrated in this work could open a way for the ultrafast all-optical control of crystal symmetry, which can find applications in frequency conversion, electro-optic modulation, electric field characterization, and so on.

Limitations of the study
In the present work, higher even-order harmonics (≥ sixth) are not detected possibly for three reasons: Firstly, based on Equation 1, the emergence of higher-order harmonics requires the THz field strength to be stronger. But here the THz field is limited by our pump infrared energy as well as the damage threshold of the DSTMS. Secondly, higher even-order harmonics observation is also limited by the spectrometer and the wavelength of the probe beam. Specifically, for the 1450-nm-centered probe light used in our experiment, the sixth harmonic is estimated to appear around 240nm, which is very close to the high-frequency limit of our spectrometer. Moreover, the signal-to-noise ratio (SNR) of measurements also plays a significant role in the search for higher-order harmonics. For the experimental results demonstrated in this work, the repetition rate of the laser system is 20 Hz. By a rough estimation, the fourth harmonic intensity is more than 4-fold than that of the sixth harmonic, which shall annihilate within the noise in our experiments. Thus, for further optimizations of current work, we expect to depict higher even-order harmonics and their relevant polarization, intensity dependence on the THz light field. In addition, more crystals can be tested beyond the centrosymmetric Si.

STAR+METHODS
Detailed methods are provided in the online version of this paper and include the following:

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ACKNOWLEDGMENTS
This work was financially supported by 100 Talents Program of CAS; Shanghai Pujiang Program (19PJ1410500), CAS scientific instruments and equipments development project (YJKYYQ20200031), National Natural Science Foundation of China (Grants No. 11922412, 11874372), Shanghai Sailing Program (Grant No.21YF1453900), Key Research Program of Frontier Sciences, CAS, grant No. ZDBS-LY-SLH018, Youth Innovation Promotion Association of Chinese Academy of Sciences, and Shanghai Pilot Program for Basic Research – Chinese Academy of Science, Shanghai Branch.

AUTHOR CONTRIBUTIONS
Conceptualization, L.S., Y.Z., and Y.D.; Formula analysis, L.S. and Y.D.; Investigation, Y.D., Y.Z., and L.S.; Resources, Z.L., J.Q., Y.P., L.S., Y.T., Y.L., and R.L.; Writing-Original Draft, Y.D.; Writing-Review & Editing, Y.Z., L.S., Y.T., and Y.D.; Project Administration, L.S. and Y.T.

DECLARATION OF INTERESTS
The authors declare no competing interests.
KEY RESOURCES TABLE

| REAGENT or RESOURCE | SOURCE | IDENTIFIER |
|---------------------|--------|------------|
| Chemicals, peptides, and recombinant proteins | | |
| DSTMS | Swiss Terahertz co | N/A |
| Silicon | Norcada co | SM1048N |
| GaP | Eachwave Scientific Instrument co. ltd | 2735085243 |
| | | |
| Other | | |
| Spectrometer | Ocean Optics Inc. | USB 4000 |
| THz CCD | Swiss Terahertz co | RIGI Camera |
| NIRCCD | Ophircopt co | SP620U |
| THz power meter | Ophir Inc. | 3A-P-THz |
| NIR power meter | Thorlabs, Inc. | PM100D |

RESOURCE AVAILABILITY

Lead contact
Further information and requests for resources and samples should be directed to and will be fulfilled by the lead contact, Liwei Song (slw@siom.ac.cn).

Materials availability
This work did not generate new unique samples.

Data and code availability

- All data reported in this paper will be shared by the lead contact upon request.
- This paper does not request original code.
- Any additional information required to reanalyze the data reported in this paper is available from the lead contact upon request.

METHOD DETAILS

Terahertz generation
The THz pulses are generated by optical rectification in an organic crystal DSTMS, and a filter is used to block the infrared pulses behind the crystal.

Electro-optic sampling method
Electro-optic sampling technique is one of the traditional techniques used to measure the THz electric field. In this work we also measured the THz electric field in this method with 50 μm-thick, (110) cut GaP crystal. Based on the Pockels effect, the index refraction of the GaP crystal is modulated when a pulsed THz radiation illuminates which is detected by a femtosecond optical probe beam. The direction and angle of these changes are proportional to the co-propagated THz pulse amplitude and phase. Combined with the use of a chopper and phase-locked amplifier, the signal-to-noise is significantly improved. The THz electric field measurement is a purely electro-optic process, and when it is velocity-matched, the system bandwidth is mainly limited by the dispersion of the THz signal, and the duration of the laser pulse in the crystal.

Laser source
The laser source is a near-infrared optical parametric chirped-pulse amplifier (OPCPA) which generates laser pulses with 1450 nm center wavelength and 120fs pulse duration at a repetition rate of 20 Hz.
Calculation of the THz electric field and IR intensity

The calculated peak value of THz electric field is 500 kV/cm, which is based on the following formula:

\[ E_{\text{THz}} = \sqrt{\frac{\epsilon_{\text{THz}}}{c\epsilon_0\tau_{\text{THz}} \pi r_{\text{THz}}^2}} \]

With \( \epsilon_{\text{THz}} \) refers to the THz pulse energy measured by the THz power meter, \( c \) refers to the speed of the light, \( \epsilon_0 \) refers to permittivity of vacuum, \( \tau_{\text{THz}} \) refers to the THz pulse duration provided by EOS, \( r_{\text{THz}} \) is the THz spot radius measured by the THzCCD.

The power density (PD) of the THz and the infrared beams on the silicon sample are 0.66 GW/cm\(^2\) and 61 GW/cm\(^2\) respectively, which are based on the following formula:

\[ PD = \frac{P_p}{\pi r^2} \]

With \( P_p \) is the peak power measured by the THz power meter and NIR power meter, and \( r \) means the spot radius measured by the THzCCD and NIRCCD.

Detection of the harmonics

In this study, the generated even-order harmonics are detected by a spectrometer (USB4000, Ocean Optics Inc.). The second and fourth harmonics are around 745 and 365 nm, and the spectrometer has a wavelength range from 200 to 1045 nm.

Calculation of the coherence length

According to the following formula:

\[ L_c = \frac{2}{|2k_1 - k_2|} \]

where \( k_1 \) and \( k_2 \) are the wave vectors of fundamental (1450 nm) and second harmonic (745 nm) in the Si sample. The coherence length is calculated to be 1.792 \( \mu \text{m} \).