Supporting Information

Laterally emitted surface second harmonic generation in a single ZnTe nanowire

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1. The calibration of the μ -PL system

As previously reported, surface SHG could also be generated on the quartz substrate when pumped by intense laser.^{1,2} Optical devices in the μ -PL system may have influence on the signal properties as well. It is necessary to calibrate of our μ -PL system in advance to rule out the influence of the quartz substrate. **Figure s1(a)** presents the measured transmitted spectrum of a blank quartz substrate with a laser pumping power of 8.2 mW. One can see clearly that no obvious SHG signal at 400 nm is observed, indicating the negligible influence of the quartz substrate. In the

experiment, the pumping laser from the mode-locked Ti/Sapphire oscillator is horizontally polarized. The polarization of the pumping laser through the optical microscope system was measured by rotating the Glan prism in front of the monochromator. **Figure s1(b)** shows the measured light intensity as a function of the signal polarization angle. One can see clearly that the largest light intensity is obtained at 0° and 180° , indicating that the 800-nm light through the optical system is still horizontally polarized. Therefore, we can conclude that the optical system does not have influence on the polarization properties of the signal.

Figure s1. (a) The measured transmitted spectrum of the blank quartz substrate with a laser pumping power of 8.2 mW. (b) Polar plot of the measured light intensity as a function of the signal polarization angle.

2. The estimation of the SHG conversion efficiency

The conversion efficiency, η , was estimated based on the ratio between the peak value of the SHG signal and that of the pumping laser, $P_{\text{SHG}}/P_{\text{pump}}$. Since only the transmitted SHG signal was detected in the experiment, the peak value of the SHG signal can be written as $P_{\text{SHG}} = \gamma P_{\text{Detect}}$, where P_{Detect} is the measured peak value of the SHG signal. γ is estimated to be 10 from the angle-resolved measurement results in

Figure 4a (assuming the equal SHG distribution on both sides of the substrate). In addition, the conversion efficiency can be further modified by considering the quantum efficiency of the PMT and some other factors through:

$$\eta = \left[\left(\frac{G_{Pump}}{G_{SHG}} \right) \cdot \left(\frac{\alpha_{Pump}}{\alpha_{SHG}} \right) \cdot \left(\frac{\beta_{Pump}}{\beta_{SHG}} \right) \cdot \left(\frac{S_{Pump}}{S_{NW}} \right) \right] \cdot \frac{\gamma P_{Detect}}{P_{Pump}}, \quad (s1)$$

where G_{Pump} and G_{SHG} are the PMT gains for the pumping laser and SHG signal; α_{Pump} and α_{SHG} are the grating efficiencies at 800 nm and 400 nm; β_{Pump} and β_{SHG} are the quantum efficiencies of the PMT at 800 nm and 400 nm; S_{Pump} is the area of the focal spot and S_{NW} is the area of the nanowire within the focal spot, respectively.

Since the SHG signal is much weaker than the pumping laser in our experiment, it requires a larger PMT gain voltage for the SHG signal than that for the pumping laser. A He-Ne laser at 632.8 nm was used to estimate the ratio of PMT gains. The peak values of the He-Ne laser were measured under the PMT gain voltages for the SHG signal and the pumping laser respectively. The ratio of PMT gains, G_{SHG}/G_{pump} , was estimated to be $\sim 1.6 \times 10^4$ in our experiment. According to the datasheets,^{3,4} α_{Pump} and α_{SHG} are estimated to be 60% and 50%, β_{Pump} and β_{SHG} are estimated to be 0.5% and 20% respectively. The diameter of the focal spot is 4 μm and then the area (S_{Pump}) is $4\pi \mu\text{m}^2$. The nanowire size is $\sim 1 \mu\text{m}$ and then the area within the focal spot (S_{NW}) is $\sim 4 \mu\text{m}^2$. All the ratio values are listed in **Table s1**.

Table s1. Related ratio values for the estimation of SHG conversion efficiency.

$G_{\text{Pump}}/G_{\text{SHG}}$	$1/(1.6 \times 10^4)$
$\alpha_{\text{Pump}}/\alpha_{\text{SHG}}$	60%/50%
$\beta_{\text{Pump}}/\beta_{\text{SHG}}$	0.5%/20%
$S_{\text{Pump}}/S_{\text{NW}}$	$4\pi/4$

Figure s2(a) shows the measured peak values of the pumping laser (P_{Pump}) and the SHG signal (P_{Detect}) as functions of the pumping power. The SHG conversion efficiency can be estimated by **Eq.(s1)**. **Figure s2(b)** shows the estimated conversion efficiency (η) as a function of the pumping power. We can see that the SHG conversion efficiencies are in the order of 10^{-6} in our experiment. In particular, conversion efficiency is estimated to be 5×10^{-6} at 8.2 mW.

Figure s2.(a) Measured peak values of pumping laser (P_{Pump}) and SHG signal (P_{Detect}) as functions of the pumping power. The ratio of the PMT gains at 400 nm and 800 nm, $G_{\text{SHG}}/G_{\text{Pump}}$, is $\sim 1.6 \times 10^4$ in the experiment. **(b)** Estimated SHG conversion efficiency, η , as a function of the pumping power.

3. Theoretical analysis based on the Gaussian fitting

Intensity distribution of the pumping beam is treated as a Gaussian profile, which

could be expressed by

$$I = I_0 \exp(-2 \frac{x^2 + y^2}{\omega_0^2}) \quad (s2)$$

When the laser beam scans through the nanowire, we need to integrate the intensity of the beam over one dimension to obtain the field distribution along the scanning line.⁵

For a nanowire oriented along the x -axis, the intensity distribution along y -axis can be written as,

$$I_r = \int_{-\infty}^{+\infty} I dx = \int_{-\infty}^{+\infty} I_0 \exp(-2 \frac{x^2 + y^2}{\omega_0^2}) dx = \sqrt{\frac{\pi}{2}} I_0 \omega_0 \exp(-2 \frac{y^2}{\omega_0^2}) \quad (s3)$$

As it has been demonstrated that the SHG power is proportional to the square of the pumping power, the SHG intensity distribution along y axis is

$$I_{SHG} \propto I_r^2 = \frac{\pi \omega_0^2 I_0^2}{2} \exp(-4 \frac{y^2}{\omega_0^2}) \quad (s4)$$

Equation (s4) indicates that the SHG intensity distribution is also a Gaussian profile.

4. Theoretical analysis of the anisotropy of SHG in the ZnTe nanowire

ZnTe has a zinc blende structure. The ZnTe nanowires grow along $[1\bar{1}1]$ direction. **Figure s3(a)** shows the geometry used to analysis the anisotropy of SHG in the ZnTe nanowire. The two coordinate systems are established on the crystal axes (xyz) and the laboratory coordinate system ($e_1 e_2 e_3$), respectively. e_1 is along the nanowire growth-axis, thus $e_1 = (1, -1, 1)$. k is wave vector of the pumping laser, which can be set as $k = (x, y, z)$. Note that $x - y + z = 0$ because k is vertical to the nanowire growth-axis (e_1). e_3 is vertical to both e_1 and k , thus it can be given by the equation

$\mathbf{e}_3 = \mathbf{k} \times \mathbf{e}_1 = (y+z, z-x, -x-y)$. The electric field, $\mathbf{E}$, is in the $\mathbf{e}_1$ - $\mathbf{e}_3$ plane and the three components in xyz system could be written as

$$\begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} = E \cos \theta \cdot \frac{1}{\sqrt{3}} \begin{bmatrix} 1 \\ -1 \\ 1 \end{bmatrix} + E \sin \theta \cdot \frac{1}{\Delta} \begin{bmatrix} y+z \\ z-x \\ -x-y \end{bmatrix} = E \begin{bmatrix} \frac{1}{\sqrt{3}} \cos \theta + \frac{y+z}{\Delta} \sin \theta \\ -\frac{1}{\sqrt{3}} \cos \theta + \frac{z-x}{\Delta} \sin \theta \\ \frac{1}{\sqrt{3}} \cos \theta + \frac{-x-y}{\Delta} \sin \theta \end{bmatrix}, \quad (\text{s5})$$

where θ is the polarization angle between $\mathbf{E}$ and $\mathbf{e}_1$, and $\Delta = [(y+z)^2 + (z-x)^2 + (-x-y)^2]^{1/2}$.

For the $\bar{4}3m$ crystal of ZnTe, there is only one independent component of the second-order nonlinear susceptibilities, $\chi_{xyz}^{(2)} = \chi_{yxz}^{(2)} = \chi_{zyx}^{(2)} = \chi$. The polarization components in xyz system are

$$\begin{aligned} \bar{P}_x &= \chi E_y E_z \hat{x} = \chi E^2 \left(-\frac{1}{3} \cos^2 \theta + \frac{1}{\sqrt{3}} \cos \theta \sin \theta \cdot \frac{y+z}{\Delta} + \frac{(x-z)(x+y)}{\Delta^2} \sin^2 \theta \right) \hat{x} \\ \bar{P}_y &= \chi E_z E_x \hat{y} = \chi E^2 \left(\frac{1}{3} \cos^2 \theta + \frac{1}{\sqrt{3}} \cos \theta \sin \theta \cdot \frac{z-x}{\Delta} + \frac{-(y+z)(x+y)}{\Delta^2} \sin^2 \theta \right) \hat{y} \\ \bar{P}_z &= \chi E_y E_x \hat{z} = \chi E^2 \left(-\frac{1}{3} \cos^2 \theta + \frac{1}{\sqrt{3}} \cos \theta \sin \theta \cdot \frac{-x-y}{\Delta} + \frac{(y+z)(z-x)}{\Delta^2} \sin^2 \theta \right) \hat{z}, \end{aligned} \quad (\text{s6})$$

where $\hat{x}, \hat{y}, \hat{z}$ are the unit direction vectors of the corresponding axes. Then the three polarization components could be decomposed and reassembled to the components parallel and vertical to the nanowire growth-axis ($\mathbf{e}_1$) in the $\mathbf{e}_1 \mathbf{e}_2 \mathbf{e}_3$ system as

$$\bar{P}_p = \frac{1}{\sqrt{3}} (|P_x| - |P_y| + |P_z|) \hat{e}_1, \quad (\text{s7})$$

$$\bar{\mathbf{P}}_s = (|\mathbf{P}_x| \cdot \frac{y+z}{\Delta} + |\mathbf{P}_y| \cdot \frac{z-x}{\Delta} + |\mathbf{P}_z| \cdot \frac{-x-y}{\Delta}) \hat{\mathbf{e}}_3. \quad (\text{s8})$$

Putting **Eq. (s6)** into **(s7)** and **(s8)** respectively, the two polarization components ($\mathbf{P}_p$ and $\mathbf{P}_s$) as functions of the angle θ can be written as

$$\bar{\mathbf{P}}_p = \frac{\chi E^2}{\sqrt{3}} (-\cos^2 \theta + \frac{1}{2} \sin^2 \theta) \hat{\mathbf{e}}_1, \quad (\text{s9})$$

$$\bar{\mathbf{P}}_s = \frac{\chi E^2}{\sqrt{3}} (\sin \theta \cos \theta - t \sin^2 \theta) \hat{\mathbf{e}}_3, \quad (\text{s10})$$

$$t = \frac{3\sqrt{3}(z-x)(x+y)(y+z)}{\Delta^3} \in [-\frac{\sqrt{2}}{2}, \frac{\sqrt{2}}{2}].$$

The SHG intensities of the corresponding components are,

$$I_p = A P_p^2 = A' (\cos^2 \theta - \frac{1}{2} \sin^2 \theta)^2, \quad (\text{s11})$$

$$I_s = B P_s^2 = B' (\sin \theta \cos \theta - t \sin^2 \theta)^2, \quad (\text{s12})$$

and the total intensity is,

$$I_T = I_p + I_s. \quad (\text{s13})$$

Figure s3(b) shows the measured results (dots) and the corresponding fittings (lines). **Figure s3(c)** shows the calculated results according to **Eq. (s11)-(s13)**. It indicates that the theoretical and experimental results are in good agreement.

Figure s3. (a) Geometry for the polarization analysis of SHG, the pumping laser was incident along $e_2(k)$. (b) Polar plot of the experimental results of the SHG intensities as functions of the polarization of the pumping laser, which are also shown in **Fig. 3(d)**. (c) Polar plot of the calculated results of the SHG intensities as functions of the polarization of the pumping laser.

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