Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Structural Discontinuity induced Surface Second Harmonic Generation in single thin Zinc-blende GaAs Nanowires

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1 Calculation of three Euler angles (α, β, γ) between the lab coordinate system and the crystal axes

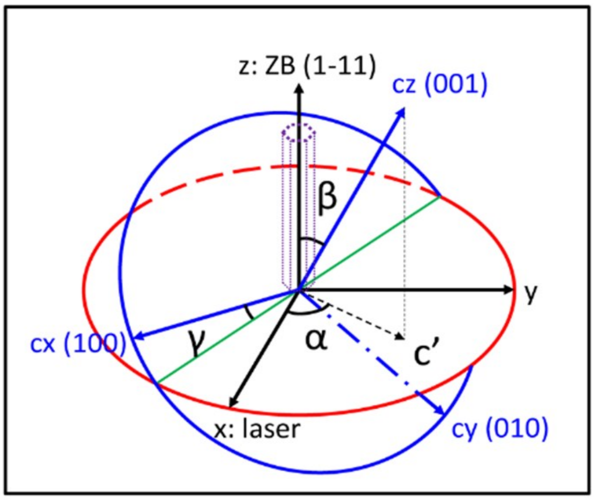

Figure S1 Geometries of the lab frame (X,Y,Z) and the crystal frame (CX,CY,CZ). Specifically, β is the angle between cz axis and z axis, α is the angle between x axis and the cz axis's projection on xy plane, and γ is the angle between cx axis and the intersection line of xy and cx-cy planes.

Note that GaAs nanowire of zinc-blende crystal structure is growing along $[1\bar{1}1]$, thus $\vec{e}_z=(1,-1,1)/\sqrt{3}$. The propagation direction of the excitation light can be set as $\vec{e}_x$

=(a,b,c), where $a^2+b^2+c^2=1$. Note that a-b+c=0 and $\vec{e}_y$ can be given by the equation $\vec{e}_y=\vec{e}_z\times\vec{e}_x=(-b-c,a-c,\ a+b)/\sqrt{3}$.

According to Euler's rotation theorem, the laboratory frame (X,Y,Z axes) can be defined by:

$$\begin{pmatrix} X \\ Y \\ Z \end{pmatrix} = R_z(-\alpha)\cdot R_x(-\beta)\cdot R_z(-\gamma)\begin{pmatrix} CX \\ CY \\ CZ \end{pmatrix} \tag{S1}$$

where $R_z(-\gamma)=\begin{pmatrix} cos(\gamma) & -sin(\gamma) & 0 \\ sin(\gamma) & cos(\gamma) & 0 \\ 0 & 0 & 1 \end{pmatrix}$, $R_x(-\beta)=\begin{pmatrix} 1 & 0 & 0 \\ 0 & cos(\beta) & -sin(\beta) \\ 0 & sin(\beta) & cos(\beta) \end{pmatrix}$, and

$$R_z(-\alpha)=\begin{pmatrix} cos(\alpha) & -sin(\alpha) & 0 \\ sin(\alpha) & cos(\alpha) & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{S2}$$

So we can get that,

$$\begin{pmatrix} a & b & c \\ \frac{-c-b}{\sqrt{3}} & \frac{a-c}{\sqrt{3}} & \frac{b+a}{\sqrt{3}} \\ \frac{1}{\sqrt{3}} & \frac{-1}{\sqrt{3}} & \frac{1}{\sqrt{3}} \end{pmatrix} = \begin{pmatrix} \cos\alpha\cdot cos\gamma - sin\alpha\cdot cos\beta\cdot sin\gamma & -\cos\alpha\cdot sin\gamma - sin\alpha\cdot cos\beta\cdot cos\gamma & si \\ \sin\alpha\cdot cos\gamma + cos\alpha\cdot cos\beta\cdot sin\gamma & -\sin\alpha\cdot sin\gamma + cos\alpha\cdot cos\beta\cdot cos\gamma & -c \\ sin\beta\cdot sin\gamma & sin\beta\cdot cos\gamma & \end{pmatrix} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

that is,

$cos\beta=\frac{1}{\sqrt{3}}$, β=54.73°, γ=135°,

$$\begin{pmatrix} a \\ b \\ c \end{pmatrix} = \begin{pmatrix} -\frac{\sqrt{2}}{2}cos\alpha-\frac{\sqrt{6}}{6}sin\alpha \\ -\frac{\sqrt{2}}{2}cos\alpha+\frac{\sqrt{6}}{6}sin\alpha \\ \frac{\sqrt{6}}{3}sin\alpha \end{pmatrix}, \quad \begin{pmatrix} -c-b \\ a-c \\ b+a \end{pmatrix} = \begin{pmatrix} \frac{\sqrt{2}}{2}cos\alpha-\frac{\sqrt{6}}{2}sin\alpha \\ -\frac{\sqrt{2}}{2}cos\alpha-\frac{\sqrt{6}}{2}sin\alpha \\ -\sqrt{2}cos\alpha \end{pmatrix} \tag{S3}$$

2 Theoretical expression of TM-polarized and TE-polarized SHG signals from Zinc-blende GaAs nanowires

The electric field, E_ω, is in the y-z plane and the three components on crystal axes (

$\vec{e}_{cx}\ \vec{e}_{cy}\ \vec{e}_{cz}$) could be written as,

$$\begin{pmatrix} E_{\omega,cx} \\ E_{\omega,cy} \\ E_{\omega,cz} \end{pmatrix} = E_\omega cos\theta_\omega \cdot \frac{1}{\sqrt{3}} \begin{pmatrix} 1 \\ -1 \\ 1 \end{pmatrix} + \gamma_{\omega,in} E_\omega sin\theta_\omega \cdot \frac{1}{\sqrt{3}} \begin{pmatrix} -c-b \\ a-c \\ b+a \end{pmatrix}$$

$$= \begin{pmatrix} \frac{1}{\sqrt{3}} cos\theta_\omega + \gamma_{w,in} \frac{-c-b}{\sqrt{3}} sin\theta_\omega \\ -\frac{1}{\sqrt{3}} cos\theta_\omega + \gamma_{w,in} \frac{a-c}{\sqrt{3}} sin\theta_\omega \\ \frac{1}{\sqrt{3}} cos\theta_\omega + \gamma_{w,in} \frac{b+a}{\sqrt{3}} sin\theta_\omega \end{pmatrix} E_\omega , \quad \text{(S4)}$$

Therefore, the parallel and perpendicular components of bulk SHG are,

$$\vec{E}^{b}_{2w,p} = \left(\vec{E}^{b}_{2w} \cdot \vec{e}_z\right)\vec{e}_z = \frac{1}{\sqrt{3}}\left(E^{b}_{2\omega,cx} - E^{b}_{2\omega,cy} + E^{b}_{2\omega,cz}\right)\vec{e}_z = \frac{2d_{14}E^2_\omega}{\sqrt{3}}\left(\frac{1}{2}\gamma^2_{\omega,in} sin^2\theta_\omega - cos^2\theta_\omega\right)\vec{e}_z ,$$

$$\vec{E}^{b}_{2w,s} = \gamma_{2\omega,out}\left(\vec{E}^{b}_{2w} \cdot \vec{e}_y\right)\vec{e}_y = \frac{2d_{14}E^2_\omega}{\sqrt{3}}\gamma_{\omega,in}\gamma_{2\omega,out}(sin\theta_\omega cos\theta_\omega - t \cdot \gamma_{2\omega,in} sin^2\theta_\omega)\vec{e}_y ,$$

$$t = (a-c)(b+a)(c+b) = \frac{\sqrt{2}(1 - 2cos2\alpha) \cdot cos\alpha}{2}$$

The TM- and TE-polarized SHG intensity becomes, $I_{2\omega,p} = A \ast \left|\vec{E}_{2\omega,p}\right|^2$ and $I_{2\omega,s} = B \ast \left|\vec{E}_{2\omega,s}\right|^2$. Thus they can be written as a function of θ_ω:

$$\begin{cases} I_{2\omega,p} = A \ast E^4_\omega\left(\frac{1}{\sqrt{3}} d_{14}\gamma^2_{\omega,in} sin^2\theta_\omega - \frac{2}{\sqrt{3}} d_{14} cos^2\theta_\omega + d^s_{zzy}\gamma_{\omega,in} sin\theta_\omega cos\theta_\omega\right)^2 \\ \qquad \propto (A_1 sin^2\theta_\omega - A_2 cos^2\theta_\omega + A_3 sin\theta_\omega cos\theta_\omega)^2 \\ I_{2\omega,s} = B \ast \gamma_{\omega,out}{}^2 E^4_\omega\left(\frac{2d_{14}}{\sqrt{3}}\gamma_{\omega,in} sin\theta_\omega cos\theta_\omega - \left(\frac{2}{\sqrt{3}} t \cdot d_{14} - d^s_{yyy}\right)\gamma^2_{\omega,in} sin^2\theta_\omega + d^s_{yzz} cos^2\theta_\omega\right)^2 \\ \qquad \propto (-B_1 sin^2\theta_\omega + B_2 cos^2\theta_\omega + B_3 sin\theta_\omega cos\theta_\omega)^2 \end{cases} \quad \text{(S5)}$$

2 Detail discussion on the relationship between the polarization and the value of $\gamma_{2w,in}$ and t(α)

TM-polarized and TE-polarized SHG is determined by field reduction factors ($\gamma_{2w,in}$, $\gamma_{2w,out}$) and the propagation direction of the incident laser beam (angle α). TM-polarized (black, $I(0^o,\theta_\omega)$) and TE-polarized (red, $I(90^o,\theta_\omega)$) as a function of excitation polarization angle (θ_ω) at different propagation directions of the incident laser beam (angle α=0^o,15^o,30^o,45^o,60^o,75^o,90^o,105^o,120^o) are shown in Figure S2 ($\gamma_{2w,in} = \gamma_{2w,out} = 1$) and S3 ($\gamma_{2w,in} = \gamma_{2w,out} = 0.5$).

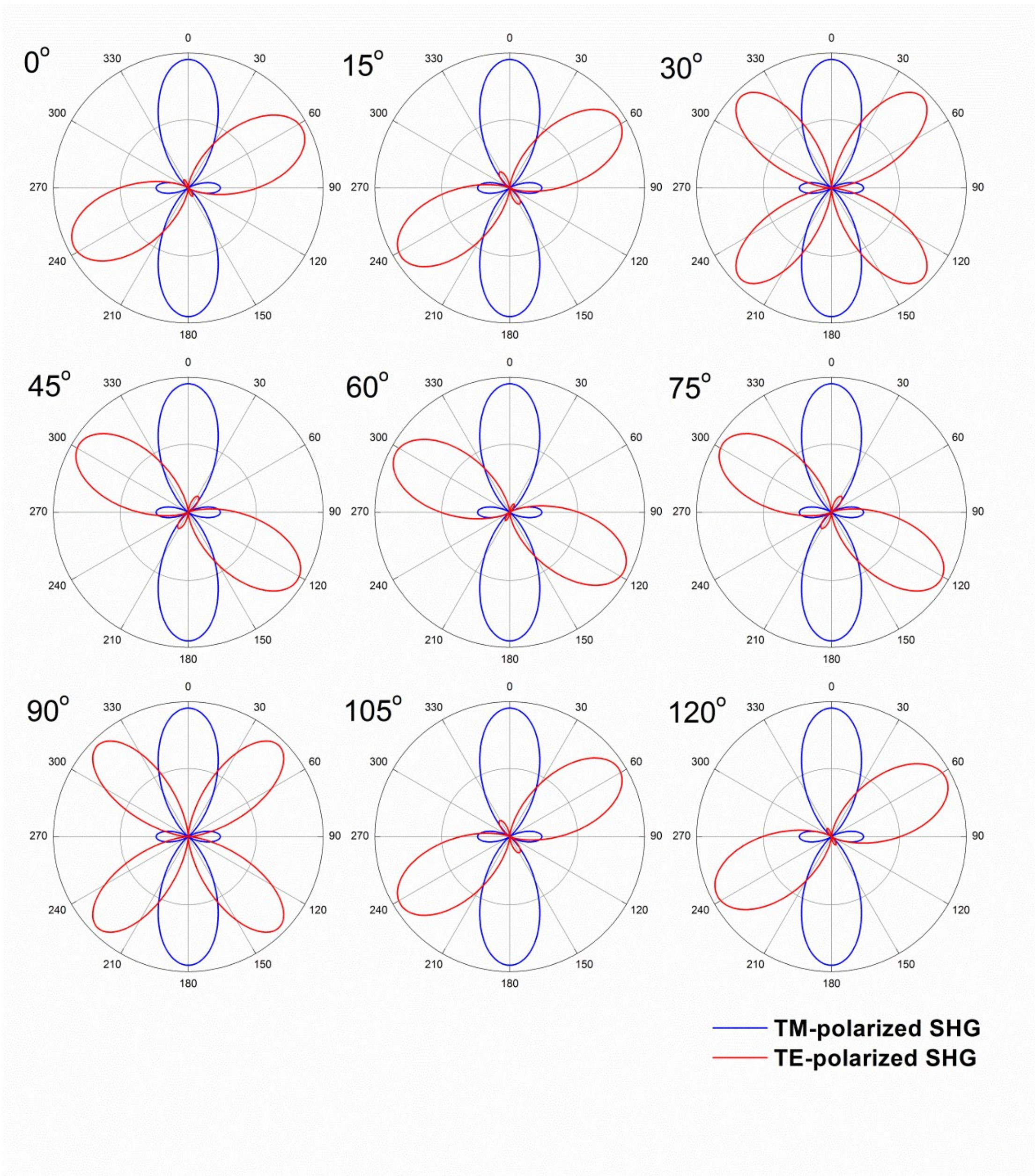

Figure S2. When $\gamma_{2w,in} = \gamma_{2w,out} = 1$, TM-polarized (blue line) and TE-polarized (red line) SHG as a function of excitation polarization angle (θ_{ω}) at different propagation directions of the incident laser beam (angle α=0°, 15°, 30°, 45°, 60°, 75°, 90°, 105°, 120°).

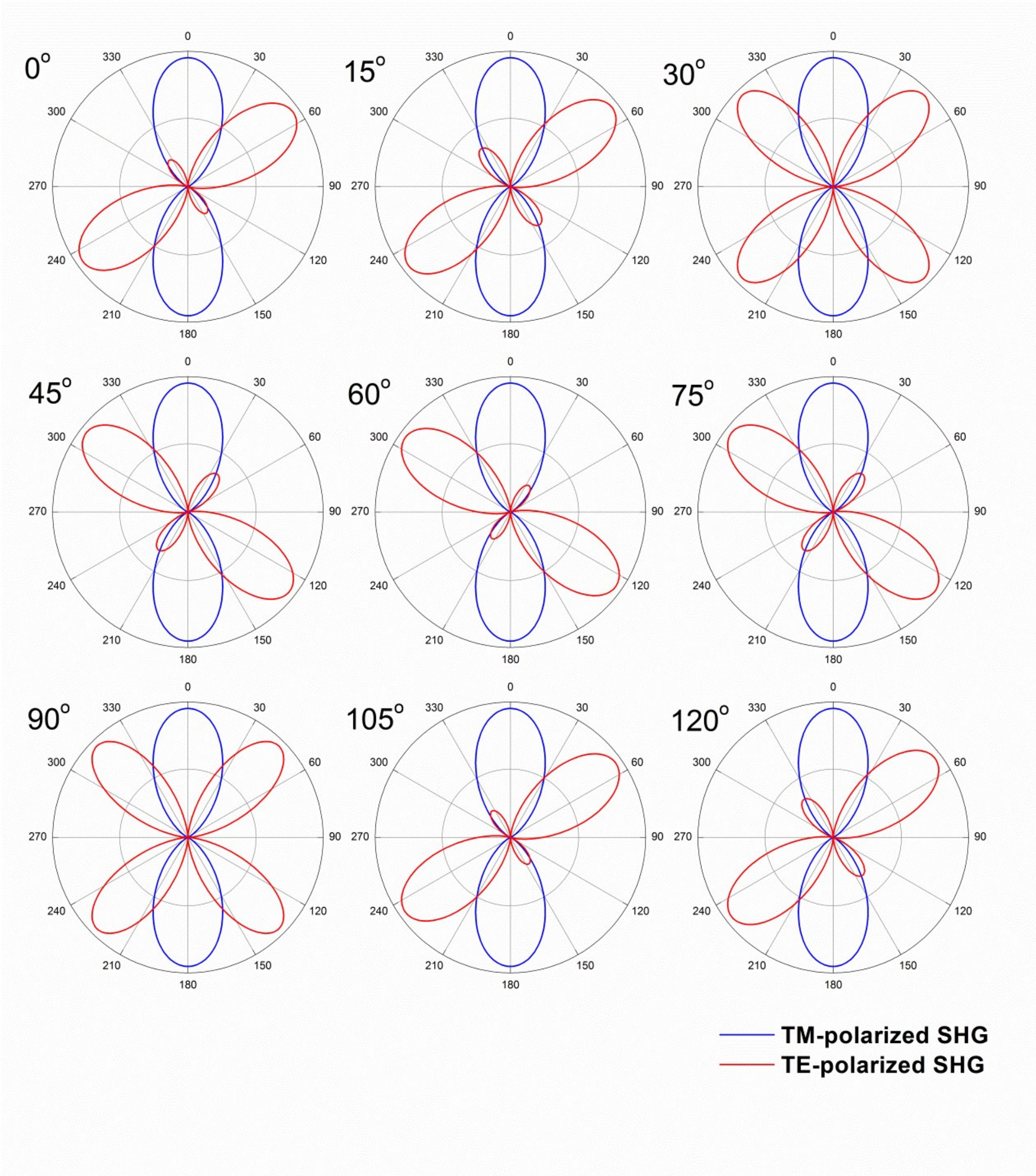

Figure S3. When $\gamma_{2w,in} = \gamma_{2w,out} = 0.5$, TM-polarized (blue line) and TE-polarized (red line) SHG as a function of excitation polarization angle (θ_{ω}) at different propagation directions of the incident laser beam (angle α=0°, 15°, 30°, 45°, 60°, 75°, 90°, 105°, 120°).