

Supplementary Information

SHG polarimetry

In order to provide an additional indication that the nature of 515nm response is indeed SHG from a wire, polarimetric studies were carried out by varying polarization of the incoming laser light. SHG intensity as a function of the pump polarization in respect to the major axis of immobilized nanowire on auxiliary wafer was analyzed. Input polarization state was controlled using half-wave plate installed on rotation mount. Results in Fig. S1 clearly demonstrate that the maximum efficiency is obtained when the light is polarized along the wire. For the perpendicular polarization the SH intensity vanishes.

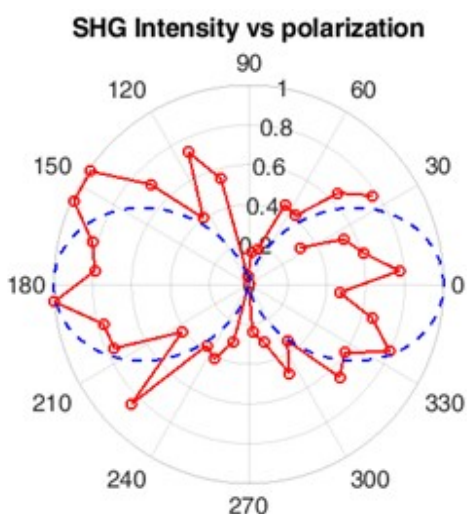


Figure S1. SHG intensity of immobilized wire versus polarization of incident laser beam. Red line – experimental data, blue line – theoretical fit.

Modulation of the second harmonic response due to Fabry-Perot NW properties

Fig. S2 demonstrates backscattered spectra with the SHG signal being modulated by the cavity.

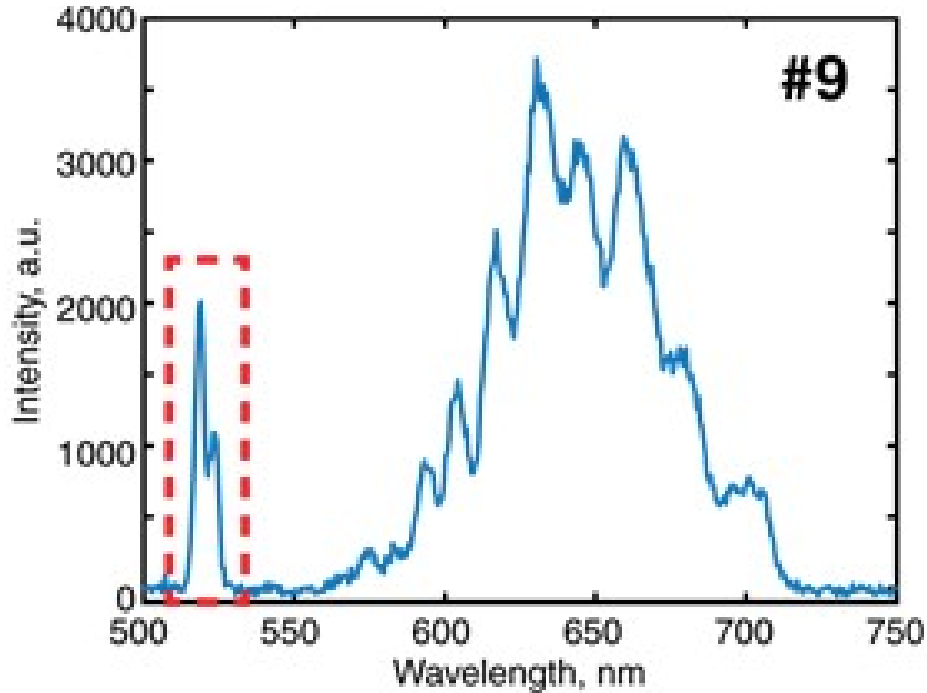


Figure S2. Back scattered spectrum from an individual GaP NW demonstrating modulation of the SHG signal due to occurrence of FP resonances.

NW length evaluation with optical microscopy and optical data analysis

To support the approach of the indirect evaluation of the NW length via analysis of the F-P modulations we trapped several NWs being firstly optically imaged in the liquid chamber drowning rather horizontally prior the trapping. Typical images are presented in Fig. S3. For correct evaluation of the nanostructures dimensions via optical imaging, camera calibration was carried out by taking images of 4 μ m silica spheres from Sigma Aldrich.

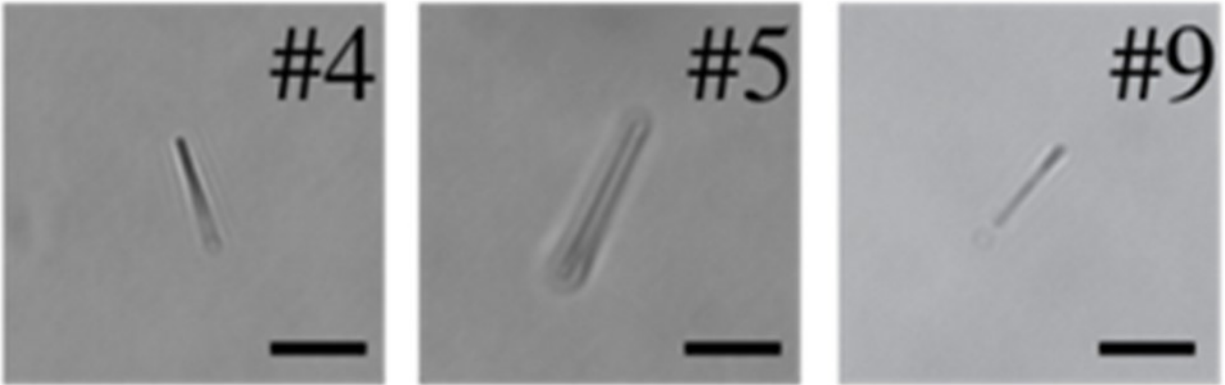


Figure S3. Optical images of individual studied nanowires. Scalebar is 5 μ m.

On the next step, scattering spectra of the same NWs were recorded and analyzed. Using the FP

formula
$$L = \frac{\lambda^2}{2\Delta\lambda(n - \lambda(dn/d\lambda))}$$
, we evaluated length of the NWs. The results of both direct optical imaging and analysis of the FP modulations are presented in Table S1.

Table S1. Summary on wire lengths obtained by image acquisition and Fabry-Perot mode spacing.

Wire #	Length by image, μ m	Length by F-P, μ m
4	6	6.6 \pm 1
5	8.7	7.5 \pm 1.5

SHG in twinned nanowires.

Here we provide a simple model to analyze components of the $\chi^{(2)}$ tensor, which correspond to a twinning structure. Changes in twinned phases do not provide a significant contribution to SHG signal and, thus, can be neglected in simulations.

The second order nonlinear tensor $\chi^{(2)}$ for zinc-blende GaP grown along [111] direction (z-axis) has the following form:

$$\begin{pmatrix} P_x^{2\omega} \\ P_y^{2\omega} \\ P_z^{2\omega} \end{pmatrix} = 2\varepsilon_0\chi^{(2)} \begin{pmatrix} d_{11} & d_{12} & 0 & 0 & d_{15}^* & d_{16} \\ d_{21} & d_{22} & 0 & d_{15}^* & 0 & d_{26} \\ d_{31}^* & d_{31}^* & d_{33}^* & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_x^\omega E_x^\omega \\ E_y^\omega E_y^\omega \\ E_z^\omega E_z^\omega \\ 2E_y^\omega E_z^\omega \\ 2E_x^\omega E_z^\omega \\ 2E_x^\omega E_y^\omega \end{pmatrix}. \quad (1.1)$$

All the elements marked with * symbol depend on the rotations along the z-axis, and thus, will take different values in each twinning grain. Their averaged value is 0, leading to

$$\langle d_{ij}^*(z) \rangle = d_{ij}^* \quad (1.2)$$

$$\langle d_{ij}(z) \rangle = 0 \quad \langle d_{ij}(z)d_{kl}(z') \rangle = \alpha^2\delta(z-z') \quad (1.3)$$

where $\alpha = d_{ij}d_{kl}l$, and l is typical thickness of single twinning layer. In the following, we will assume that $l \ll \lambda$ and on the system's scales there are many independent realizations.

Let us consider a problem of a single wave $\mathbf{E}^\omega = [b_2, 0, b_1]$ at the fundamental frequency ω propagating in 1D. This pump field is converted to the SH signal at 2ω $\mathbf{E}^{2\omega} = [a(z), 0, 0]$. The conversion process can be summarized with the following differential equation [1]:

$$\frac{d}{dz}a(z) = id_{11}(z)k_1b_1^2e^{i(2k_1-k_2)z} + id_{12}(z)k_1b_1b_2e^{i(2k_1-k_2)z}. \quad (1.4)$$

Here k_1 and k_2 are the propagation constants of the fundamental and second harmonic fields. In this model we also assume that $d_{11} = \text{const}$ along the propagation direction, while d_{12} randomly changes as discussed above. In the undepleted pump approximation b_1 and b_2 are constants, leading to:

$$a(z) = id_{11}k_1b_1^2 \int_0^z e^{i\Delta ks} ds + ik_1b_1b_2 \int_0^z d_{12}(s)e^{i\Delta ks} ds, \quad (1.5)$$

where $\Delta k = 2k_2 - k_1$ is the phase mismatch. The SH field intensity is then derived as follows:

$$\begin{aligned} |a(z)|^2 = d_{11}^2k_1^2|b_1|^4 \int_0^z \int_0^z e^{i\Delta k(s-s')} ds ds' + 2 \text{Re} \left(d_{11}k_1^2b_1^2b_2^*b_2^* \int_0^z \int_0^z d_{12}(z)e^{i\Delta k(s-s')} \right) \\ + k_1^2|b_1|^2|b_2|^2 \int_0^z \int_0^z d_{12}(s)d_{12}(s')e^{i\Delta k(s-s')} ds ds' \end{aligned} \quad (1.6)$$

Now, one should average the obtained expression over the random crystalline structure of the material taking into account that $\langle d_{12}(s) \rangle = 0$ and $\langle d_{12}(s)d_{12}(s') \rangle = \tilde{d}^2 l \delta(s - s')$:

$$\langle |a(z)|^2 \rangle = d_{11}^2 k_1^2 |b_1|^4 \int_0^z \int_0^z e^{i\Delta k(s-s')} ds ds' + k_1^2 |b_1|^2 |b_2|^2 \tilde{d}^2 l \int_0^z \int_0^z \delta(s-s') e^{i\Delta k(s-s')} ds ds' = \quad (1.7)$$

$$= \underbrace{d_{11}^2 k_1^2 |b_1|^4 \int_0^z \int_0^z e^{i\Delta k(s-s')} ds ds'}_{\text{coherent ph. match.}} + \underbrace{k_1^2 |b_1|^2 |b_2|^2 \tilde{d}^2 l z}_{\text{random ph. match.}}. \quad (1.8)$$

If the phase matching condition is fulfilled then $\Delta k = 0$ and the first term can be reduced:

$$I_{\text{SHG}}(z) \sim \langle |a(z)|^2 \rangle = d_{11}^2 k_1^2 |b_1|^4 z^2 + k_1^2 |b_1|^2 |b_2|^2 \tilde{d}^2 l z. \quad (1.9)$$

From the analysis of the last term, one can conclude that the contribution of the random phase matching is proportional into the total SHG signal is of the order $l/z \ll 1$ smaller than from the coherent phase matching term. Thus, in the analyzis of SHG emission for the random media the random components of $\chi^{(2)}$ tensor can be neglected if $\Delta k \ll 1/l$.

References:

- [1] E. Y. Morozov, A. A. Kaminskiĭ, A. S. Chirkin, and D. B. Yusupov, "Second optical harmonic generation in nonlinear crystals with a disordered domain structure," JETP Lett. 73(12), 647–650 (2001).