Resolving Radial Composition in Polarized Confocal Raman Spectra of Individual 3C-SiC Nanowires

Supporting information

The Raman scattering cross section is proportional to the time autocorrelation of the target polarizability $P_{\alpha\beta}$ and to the product of the (complex) incident and scattered electric fields $\vec{E}_{L,S}$:

$$\begin{aligned}\sigma(\omega) \propto \frac{1}{2\pi V}\int dt e^{iwt}\int d^3r\int d^3r'\int \frac{d^3q}{(2\pi)^3}\, e^{i\vec{q}\cdot(\vec{r}-\vec{r}')}\times < P_{\alpha\beta}(t)P_{\gamma\delta}(0) > \\ \times E_{\alpha}^{L}(\vec{r},\omega_L)E_{\beta}^{*S}(\vec{r}',\omega_S)E_{\gamma}^{*L}(\vec{r}',\omega_L)E_{\delta}^{S}(\vec{r}',\omega_S).\end{aligned} \tag{1}$$

The normal modes of the radiation field inside a dielectric cylinder of radius *a* and dielectric function ε are labelled as transverse magnetic (TM or E; electric field parallel to the cylinder axis) and transverse electric (TE or H; electric field perpendicular to the cylinder axis). With the incident light polarized TM, polarized and depolarized Raman cross sections are measured by analyzing the polarization of the scattered light (TM=polarized, TE=depolarized).

The normal mode expansions of the radiation field inside a cylinder are given by the following formulas, valid for normal incidence. Inside the wire:

$$\vec{E}^{TM} = \hat{z}E_0\sum_{-\infty}^{\infty} i^m b_m^E J_m(\sqrt{\varepsilon}kr)e^{im\theta} \tag{2}$$

$$\vec{E}^{TE} = E_0\sum_{-\infty}^{\infty} i^m b_m^H \left(-\frac{m}{\varepsilon kr}J_m(\sqrt{\varepsilon}kr)\hat{r} - \frac{i\sqrt{\varepsilon}}{\varepsilon}J_m^{'}(\sqrt{\varepsilon}kr)\hat{\theta}\right)e^{im\theta}, \tag{3}$$

where the coefficients are given by:

$$\begin{aligned} b_m^{E,H} &= \frac{2i/\pi ka}{J_m(\sqrt{\varepsilon}ka)H_m^{'}(ka) - \eta^{E,H}H_m(ka)J_m^{'}(\sqrt{\varepsilon}ka)}, \\ \eta^{\mathrm{E}} &\equiv \varepsilon^{1/2}, \quad \eta^{\mathrm{H}} \equiv \varepsilon^{-1/2}, \end{aligned} \tag{4}$$

with J_m, H_m Bessel and Hankel functions of the first kind (prime denoting differentiation with respect to the argument), and the $\hat{z}$-axis points along the nanowire. Analogously, outside the wire, the normal modes are:

$$\vec{E}^{TM} = \hat{z}E_o \sum_{-\infty}^{\infty} i^m \left(J_m(kr) + a_m^E H_m(kr) \right) e^{im\theta} \tag{5}$$

$$\vec{E}^{TE} = -E_0 \sum_{-\infty}^{\infty} i^m \left[\left(\frac{m}{kr} J_m(kr)\hat{r} + iJ_m^{'}(kr)\hat{\theta} \right) + a_m^H \left(\frac{m}{kr} H_m(kr)\hat{r} + iH_m^{'}(kr)\hat{\theta} \right) \right] e^{im\theta}, \tag{6}$$

$$a_m^{E,H} = \frac{\eta^{E,H} J_m(ka) J_m^{'}(\sqrt{\varepsilon}ka) - J_m^{'}(ka) J_m(\sqrt{\varepsilon}ka)}{J_m(\sqrt{\varepsilon}ka) H_m^{'}(ka) - \eta^{E,H} H_m(ka) J_m^{'}(\sqrt{\varepsilon}ka)} \tag{7}$$

Figure S1 shows the ratio of polarized to depolarised cross sections calculated neglecting Raman tensor anisotropy (for the zincblende crystal structure, this anisotropy further enhances the ratio by a factor of 4). Figure S2 shows a comparison between the intensities of the backscattered radiation field in the TM and TE polarization. Calculations assume the dielectric constant of SiC at 633 nm wavelength, ε=6.55.

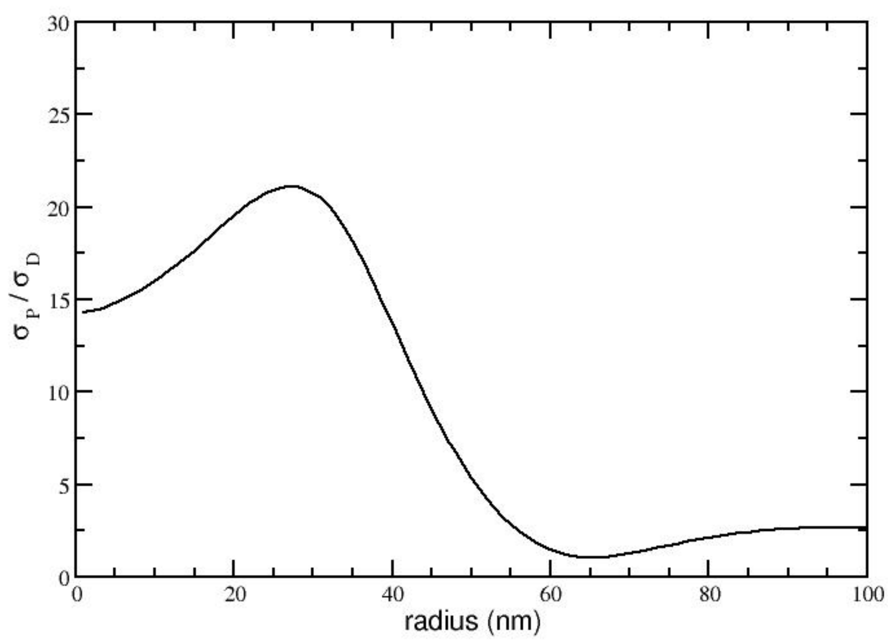

Figure S1. Ratio of the polarized to depolarized cross sections as a fuction of nanowire radius.

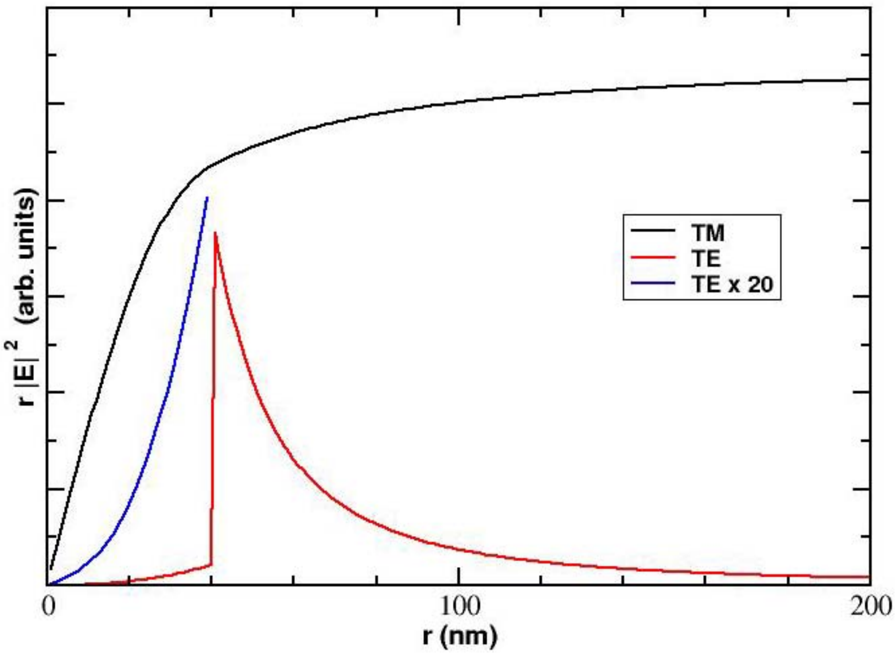

Figure S2. Amplitude squared of the TM and TE scattered modes for a 40 nm wire as a function of the distance from the wire axis

Comparison of the intensities of loss (Stokes) and gain (antiStokes) signals provides a measure of the sample temperature according to the formula:

$$\frac{\sigma_{Stokes}}{\sigma_{AntiStokes}} = \exp\left(\frac{\hbar\omega}{k_B T}\right) \tag{8}$$

Figure S3 shows the Stokes and anti-Stokes spectra from a SiC NW obtained with a 13.5 mW incident laser power. (The *x*-axis is understood to mean energy gain for the antiStokes spectrum). From the relative intensity, the sample temperature is estimated at 375 K. This relatively small temperature rise is indicative of good thermal contact with the gold substrate. We have observed that experiments on NWs suspended on a TEM grid must be conducted with much lower incident laser power (of order 100 microW) to avoid vaporization of the sample.

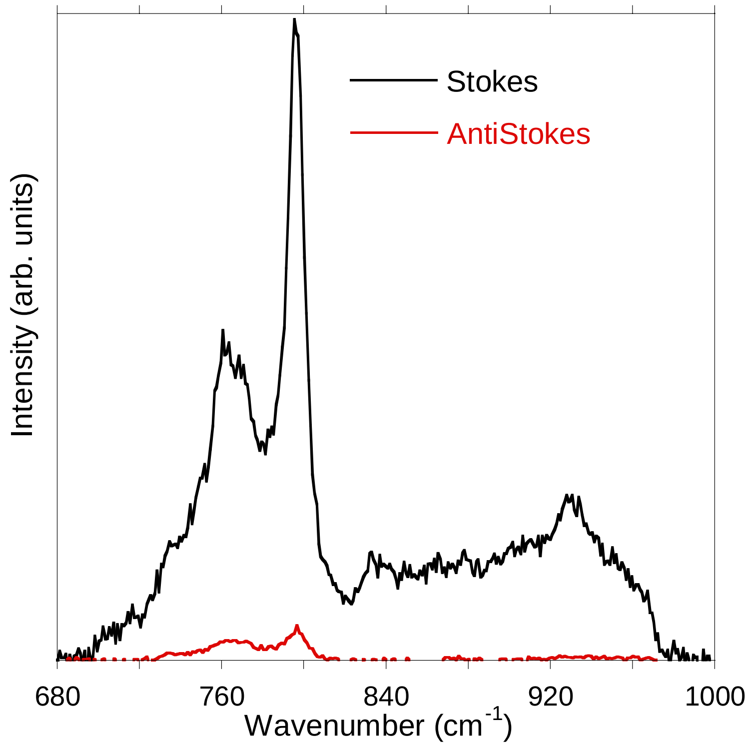

Figure S3. Comparison of the Stokes and anti-Stokes spectra for a laser power of 13.5mW. For the antiStokes spectrum, the x-axis represents energy gain rather than energy loss.