Light Manipulation and Applications
https://doi.org/10.70401/lma.2026.0012

Supplementary information

Terahertz-driven manipulation of surface-to-surface second-harmonic interference in zinc oxide

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Energy–momentum matching analysis for the PhP-assisted SHG process

The energy–momentum matching of phonon polaritons (PhPs)-assisted second harmonic generation (SHG) is characterized by dispersion relations $\omega_{\mathrm{PhP}}(k_{\mathrm{PhP}})$ and ω_{IR}. During the SHG process of infrared (IR) pulses, PhPs can be either absorbed or emitted, giving rise to Stokes and anti-Stokes scattering. Efficient nonlinear conversion requires the simultaneous conservation of energy and momentum, which determines the allowed relation among the PhP wavevector k_{PhP}, the incident IR wavevector, and the generated second harmonic wavevector. For completeness, four cases are considered in the calculation: Stokes and anti-Stokes channels with forward- and backwardpropagating PhPs. Both the upper and lower PhP branches are evaluated over a sufficiently broad frequency window to identify all possible matching solutions. The polariton dispersion, i.e., the relationship between wavevector k and frequency ω, is given by the following formula:

$$\omega_{\mathrm{up/down}}(k) = \sqrt{\frac{k^2+\omega_{\mathrm{TO}}^2\varepsilon_0}{2\varepsilon_\infty}\left(1 \pm \sqrt{1-\frac{4k^2\varepsilon_\infty\omega_{\mathrm{TO}}^2}{\left(k^2+\omega_{\mathrm{TO}}^2\varepsilon_0\right)^2}}\right)} \tag{1}$$

In the formula, ω denotes the frequency of the polariton; k denotes the wave number of the polariton k_{PhP}; ω_{TO} is the transverse optical phonon frequency, which is 410 cm^{-1}[1]; ε_0 and ε_∞ are the static and high- frequency dielectric constants, respectively, with values of 7.77 and 3.7[2]. Given a specific phonon frequency ω_{PhP}, one needs to find the corresponding wave number k_{PhP} that satisfies either $\omega_{\mathrm{up}}(k_{\mathrm{PhP}}) = \omega_{\mathrm{PhP}}$ or $\omega_{\mathrm{down}}(k_{\mathrm{PhP}}) = \omega_{\mathrm{PhP}}$.

After determining the polariton wavevector k_{PhP}, via the PhP dispersion relation, it is substituted into the energy-momentum phase-matching equations to solve for the IR and second-harmonic frequencies that satisfy the phase-matching conditions. In comparison with experimental results, we primarily consider the dispersion branch corresponding to the 1,450 nm probe wavelength employed in the measurements.

Considering the momentum and energy conservation conditions for forward- and backwardpropagating PhPs. For the forward-propagating case:

$$\omega_{\mathrm{SHG}} = 2\omega_{\mathrm{IR}} \pm \omega_{\mathrm{PhP}} \tag{2a}$$

$$k_{\mathrm{SHG}} = 2k_{\mathrm{IR}} \pm k_{\mathrm{PhP}} \tag{2b}$$

and backward-propagating:

$$\omega_{\mathrm{SHG}} = 2\omega_{\mathrm{IR}} \pm \omega_{\mathrm{PhP}} \tag{3a}$$

$$k_{\mathrm{SHG}} = 2k_{\mathrm{IR}} \mp k_{\mathrm{PhP}} \tag{3b}$$

Substituting the refractive index, and the wavevector expression $k(\omega) = n(\omega)\omega/c$ into the PhP dispersion relation and the conservation conditions, the wavevector of the second harmonic associated with Stokes scattering is obtained as:

$$k_{\mathrm{s}} = n(2\omega_{\mathrm{IR}} - \omega_{\mathrm{PhP}})\cdot(2\omega_{\mathrm{IR}} - \omega_{\mathrm{PhP}})/c \tag{4}$$

where the refractive index is described by the Sellmeier equation $n^2(\lambda) = 2.81418 + \frac{0.87968\lambda^2}{\lambda^2 - 0.3042^2} - 0.00711\lambda^2$, in order to compute the phase-matching branch structures at different photon energies. For a finite phase mismatch Δk_{s}, the corresponding coherence length is defined as: $L_{\mathrm{c}} = \pi/|\Delta k_{\mathrm{s}}|$. The term Δk_{s} denotes the phase mismatch of the Stokes channel, which is defined as the total incident wavevector minus the outgoing Stokes optical wavevector. Considering the forward- and backward-propagating configurations, the corresponding relations can be expressed as:

$$\Delta k_{\mathrm{s}}^{+} = 2k_{\mathrm{IR}} - k_{\mathrm{PhP}} - k_{\mathrm{s}} \tag{5a}$$

$$\Delta k_{\mathrm{s}}^{-} = 2k_{\mathrm{IR}} + k_{\mathrm{PhP}} - k_{\mathrm{s}} \tag{5b}$$

The wavevector k_{PhP} is derived from Eq (1). The magnitude of Δk_{s} obtained from numerical calculation can be used to judge whether the phase-matching condition is satisfied. In the main text, we substitute the experimental measured data into the formula and obtain the coherence length satisfying: $L_{\mathrm{c}} \gg d$. Accordingly, a good phase-matching condition can be achieved. For the anti-Stokes channel, the corresponding relations are obtained by replacing $2\omega_{\mathrm{IR}} - \omega_{\mathrm{PhP}}$ with $2\omega_{\mathrm{IR}} + \omega_{\mathrm{PhP}}$, and k_{s} with k_{as}. We adopt natural units in which the speed of light is set to 1, so that frequency and wavenumber have the same dimension. In numerical calculations, all frequencies and wavenumbers are taken in units of cm^{-1}, and we set $c = 1$.

The calculated complete phase-matching relationship data corresponding to the second harmonic of the probe beam are shown in Figure S1.

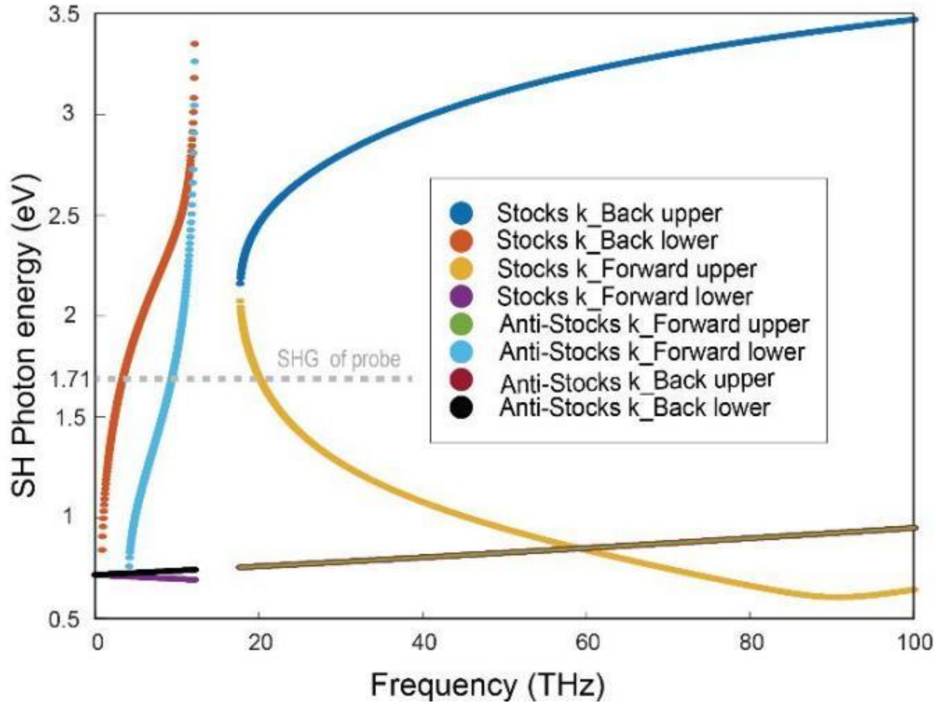

Figure S1. Calculated energy–momentum matching curves for the PhP-assisted SHG process under different matching channels. The gray dashed line represents the SHG photon energy of 1.71 eV corresponding to the experimentally used 1,450 nm probe, while the solid circles represent different phase-matching modes. PhP: phonon polariton; SHG: second harmonic generation; SH: second harmonic.

References

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