Real-Space Observation of Ultrastrong Coupling Between Optical Phonons and Surface Plasmon Polaritons

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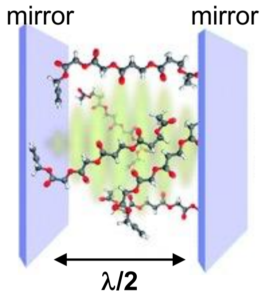
Strong coupling

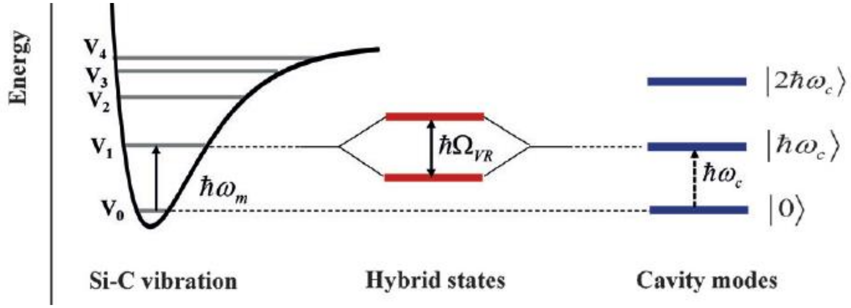
Two systems interact so strongly that they exchange energy faster than they lose it to their environments. g>losses

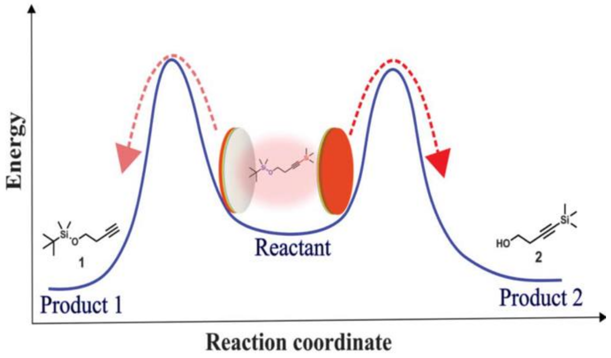
→ Change of excitation energy. $E_1, E_2 \rightarrow E_+, E_-$.

Vibrational strong coupling



Molecules in IR microcavity



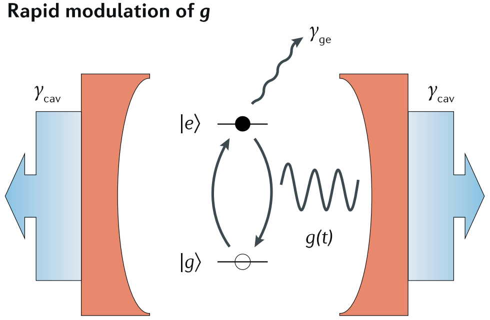


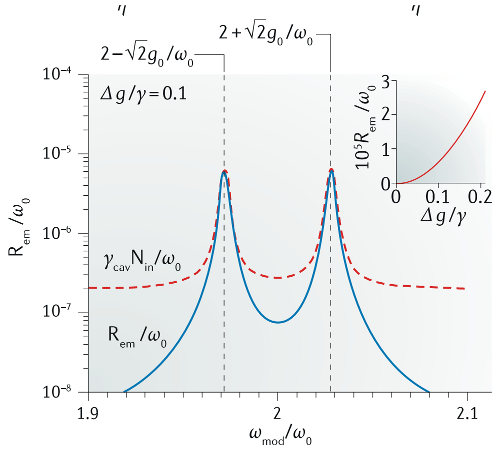
VCS can change chemical reactivity landscapes

Ultra-strong coupling

Energy exchange faster than oscillator dynamics . $g > 0.1\omega$

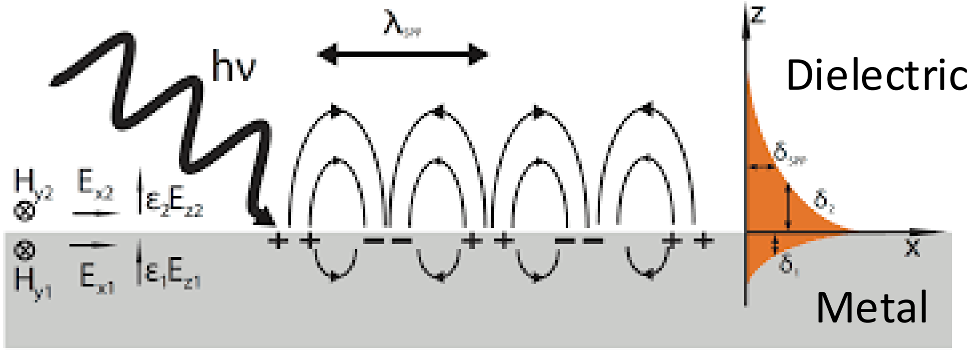
Ground state modification.



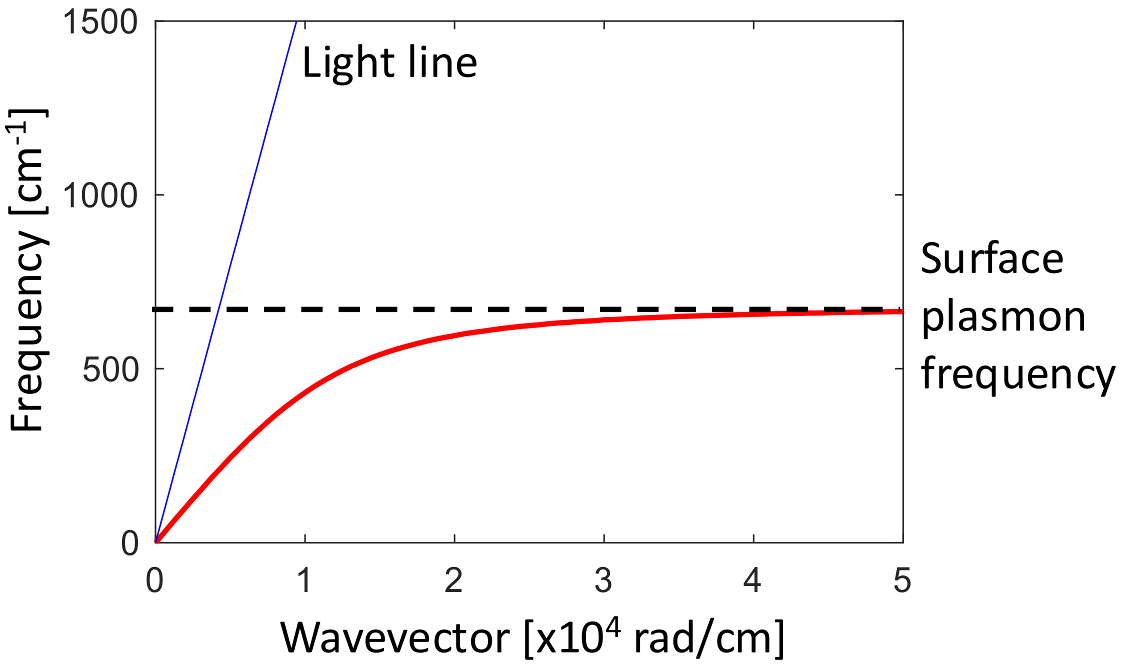


T. Ebbesen, *Acc. Chem. Res*. **49**, 2403 (2016)
A. Thomas et al., Angew. Chem. Int. **55**, 11462 (2016)
A. Thomas et al., *Science* **363**, 615 (2019)
F. Kockum. *Nat Rev Phys* **1**, 19 (2019)

Surface plasmon polariton is the consequence of light-matter coupling



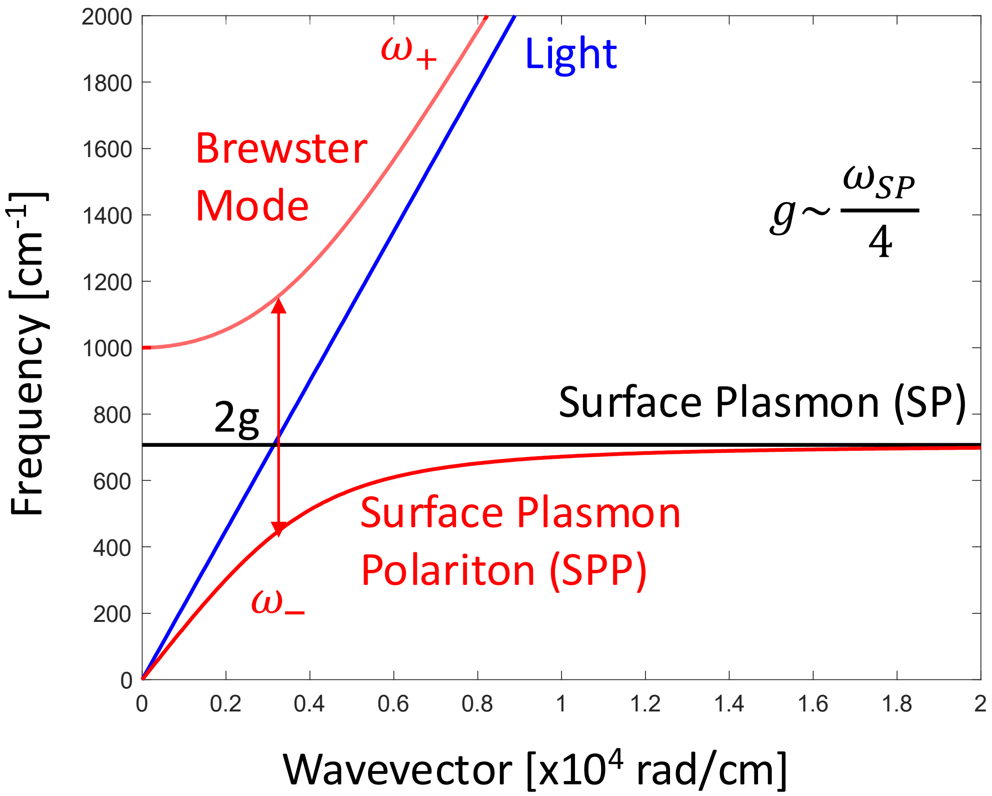
Surface Plasmon polariton is coupling between light and matter.



$k = 1/\lambda$

Surface plasmon polariton is the consequence of ultra-strong light-matter coupling

SPP dispersion

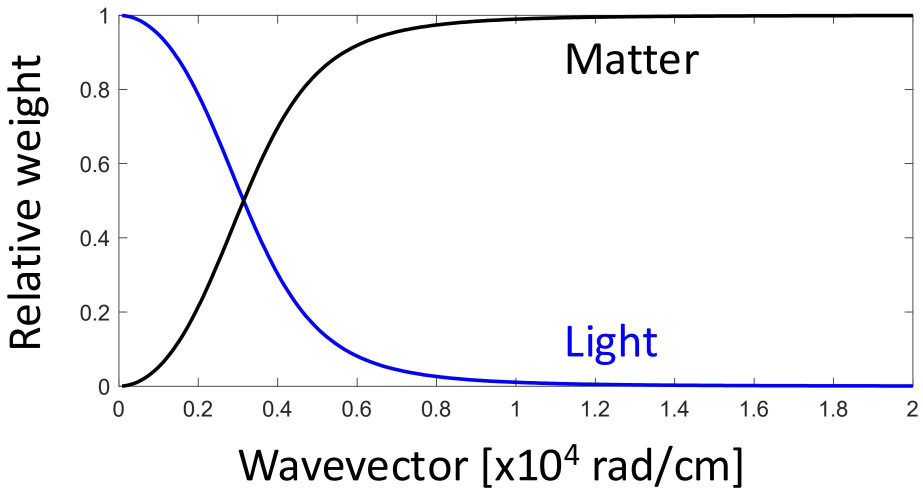


Coupled harmonic oscillator model

$$\ddot{x}_1 + \omega_{SP}^2 x_1 - 2g\dot{x}_2 = 0$$
$$\ddot{x}_2 + \omega_{light}^2 x_2 + 2g\dot{x}_1 = 0$$

$$\omega_\pm = \frac{1}{\sqrt{2}}\sqrt{\left(\omega_{SP}^2 + \omega_{light}^2 + 4g^2\right) \pm \sqrt{\left(\omega_{SP}^2 + \omega_{light}^2 + 4g^2\right)^2 - 4\omega_{SP}^2\omega_{light}^2}}$$

SPP composition

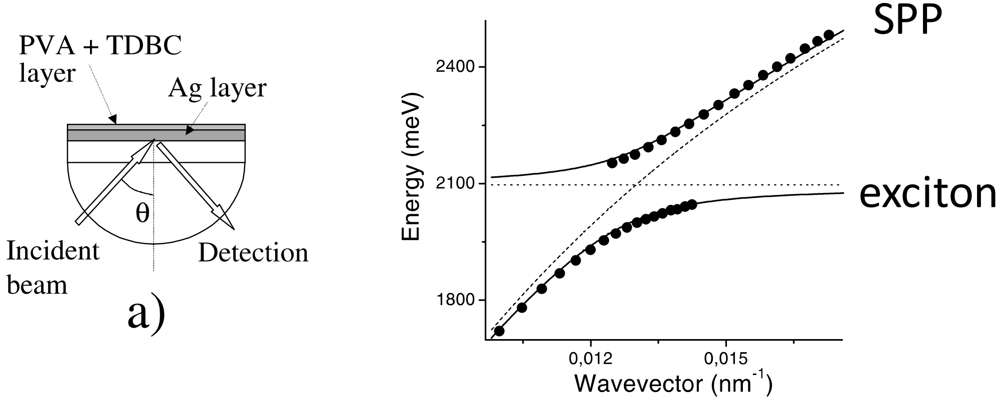


U. Muniain, *Nanophotonics*. 2025.

Strong Coupling of surface plasmon polariton with …

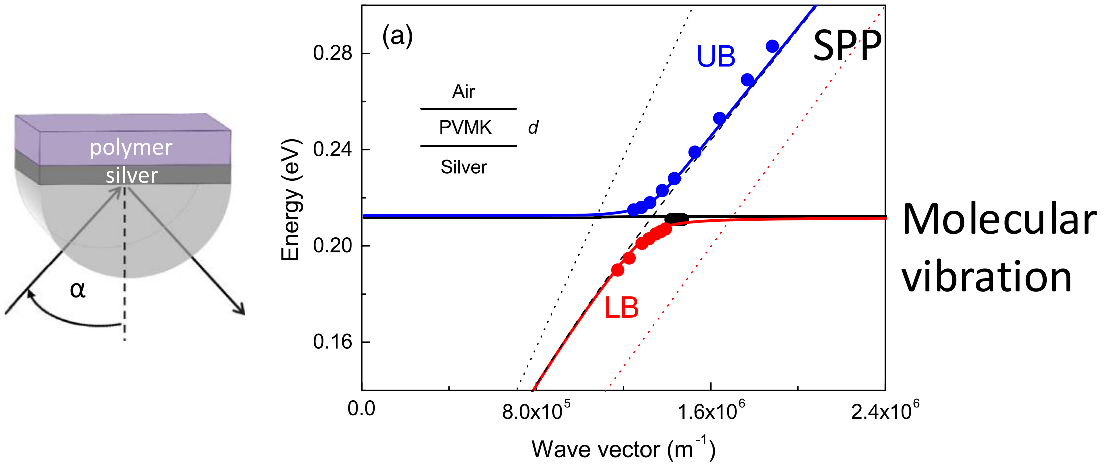
Surface plasmon polaritons have been used to achieve strong coupling with excitons or molecular vibrations thanks to its high confinement ($g \propto \sqrt{V_{overlap}}$).

Excitons



J. Bellessa Phys. Rev. Lett. 93, 03640 2004

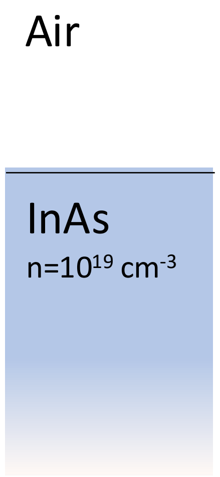
Molecules



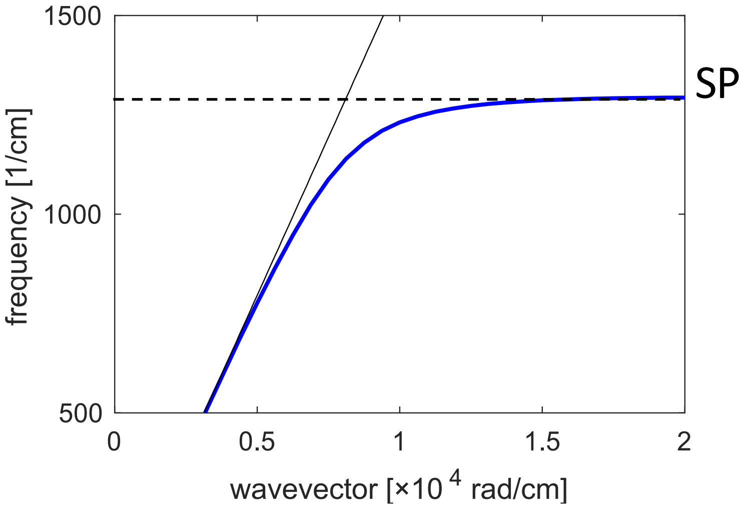
H. Memmi, Phys. Rev. Lett. 118, 126802 2017

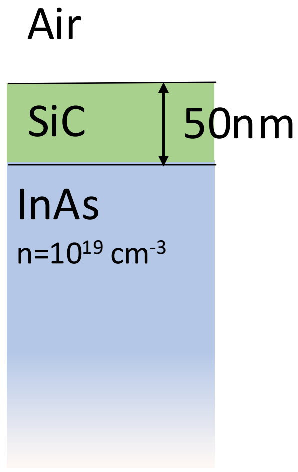
But few study of strong coupling with phonons…

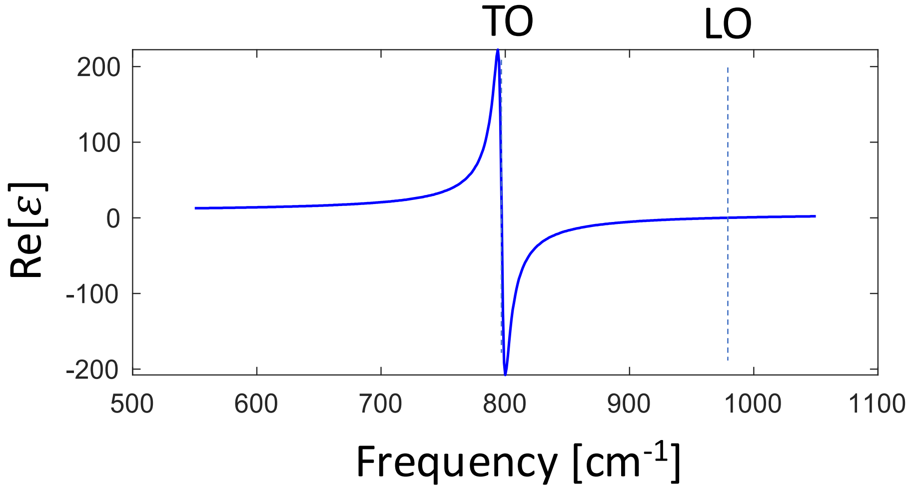
Coupling InAs's SPP with SiC phonons



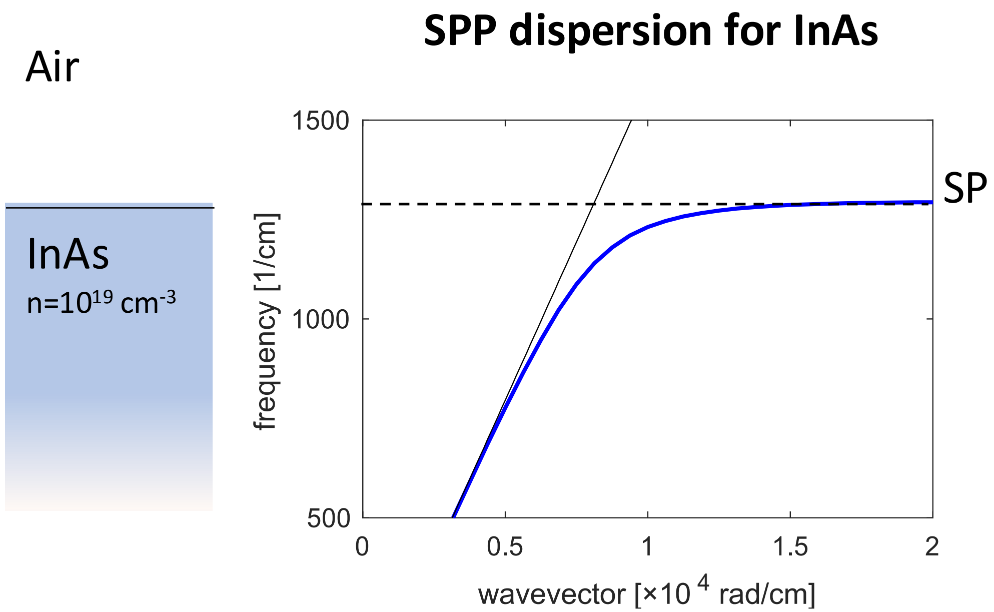
SPP dispersion for InAs

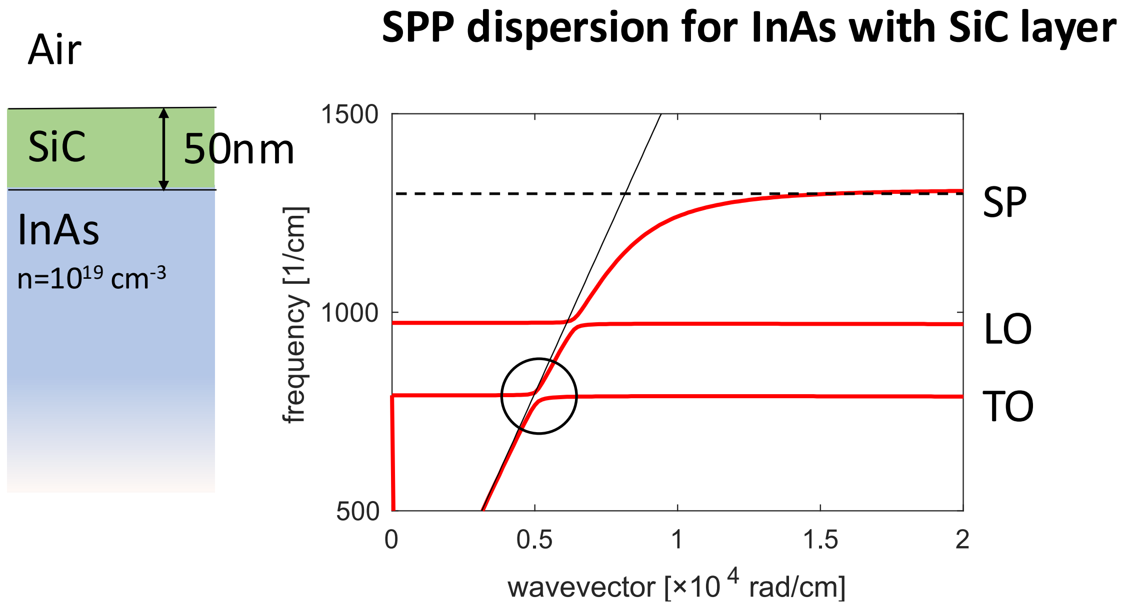






Coupling InAs's SPP with SiC transverse optical phonons

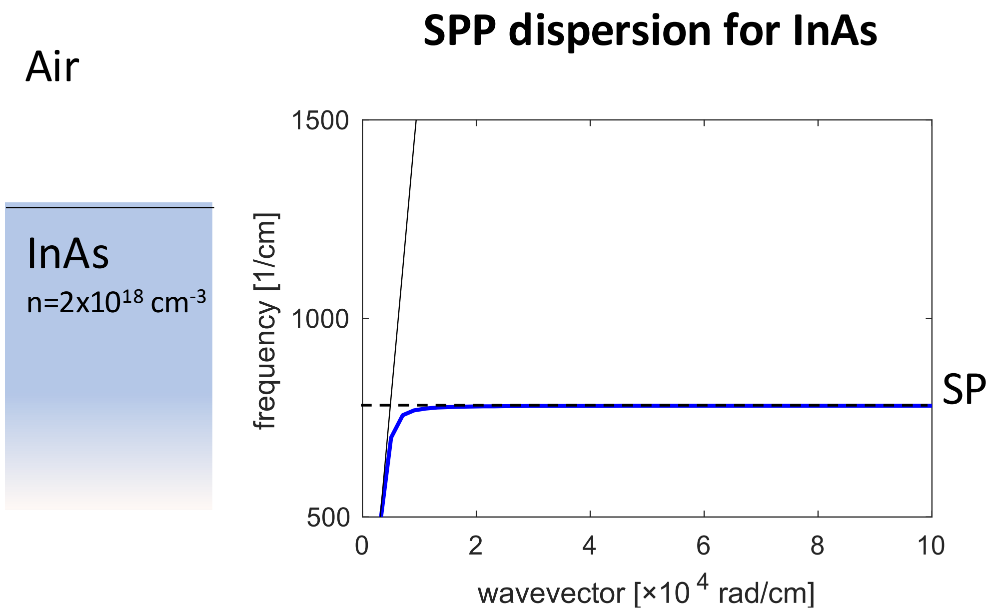


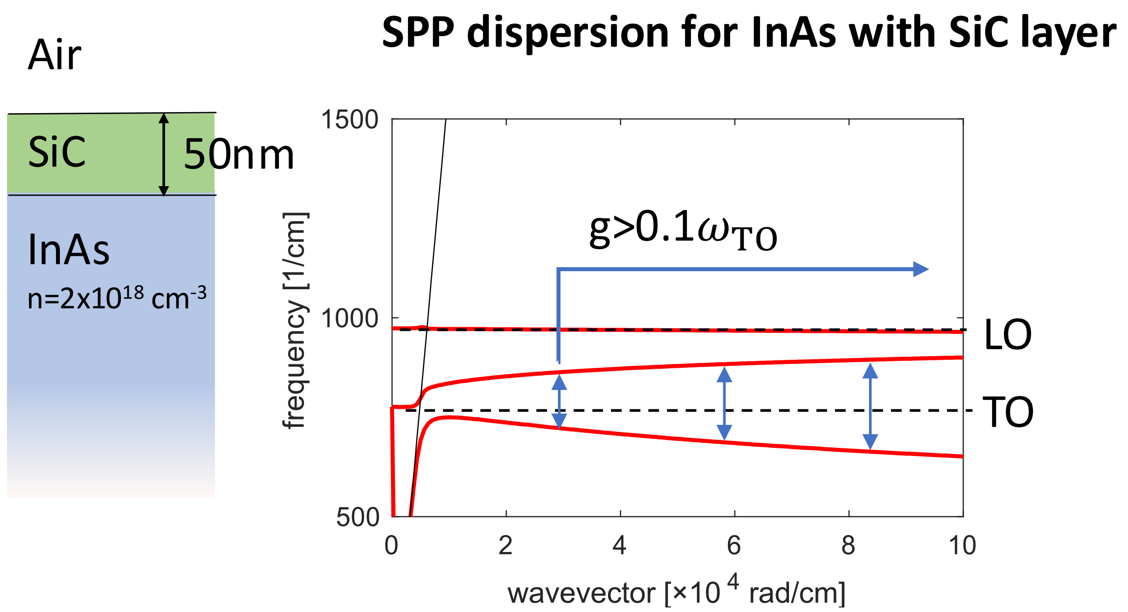


- Anti-crossing at SiC TO phonons frequency.
- Weak coupling due to low confinement of SPP.

$$g \propto \sqrt{V_{overlap}}$$

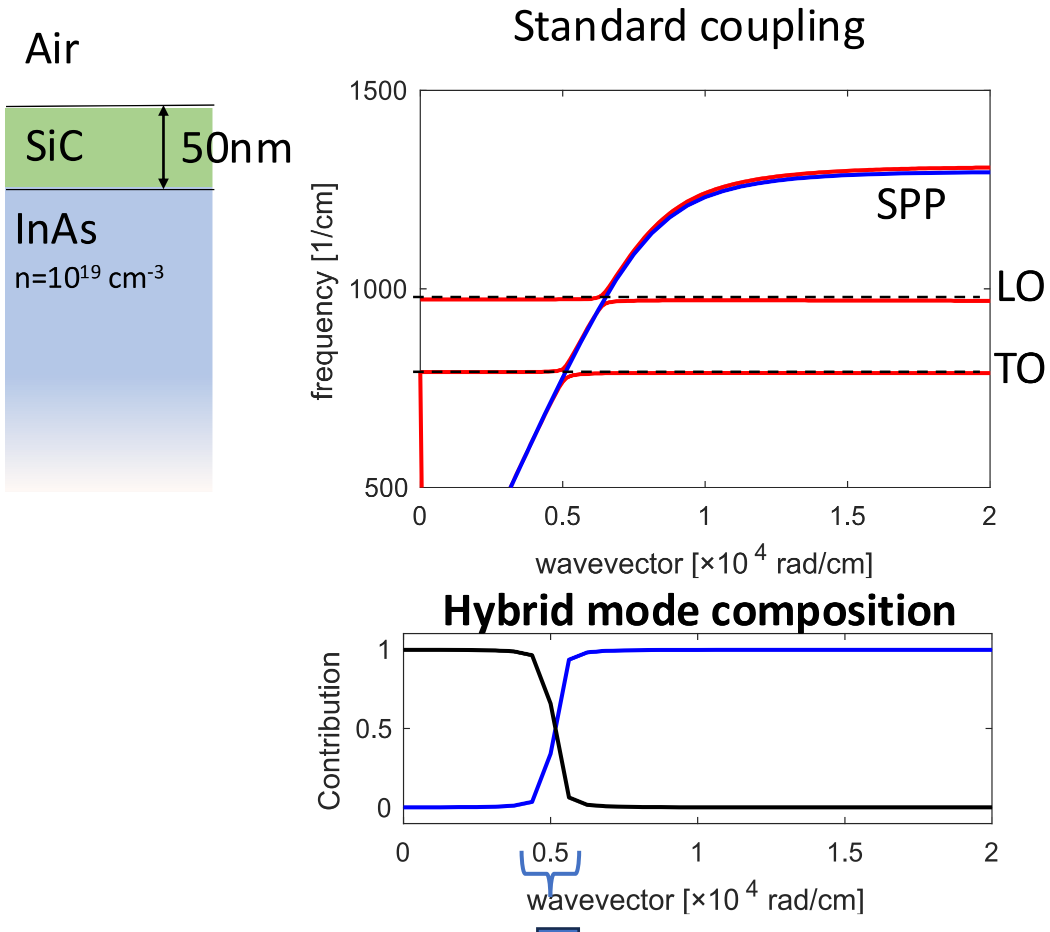
Coupling SPP dispersion limit with SiC TO phonon



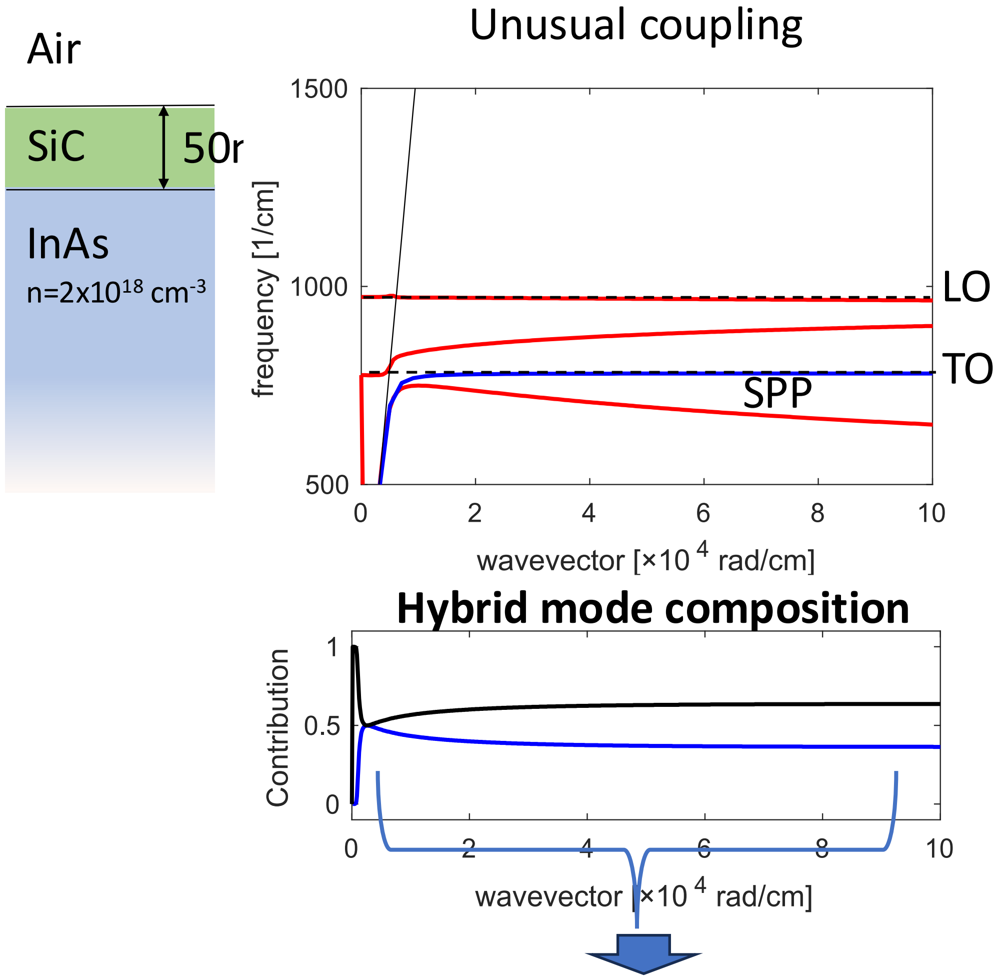


Moving SP to match TO, we identify an unusual coupling.
Ultrastrong coupling between SPP and TO phonon.

Coupling SPP dispersion limit with SiC TO phonon



mixed polariton–phonon state in a narrow wavevector range

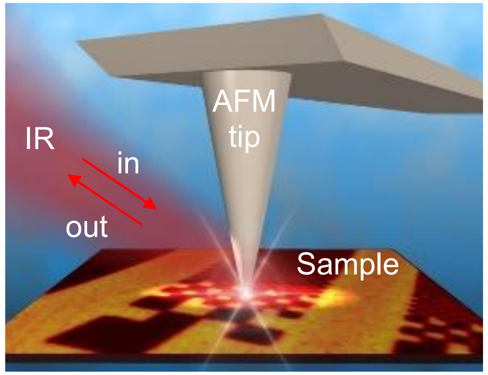


mixed polariton–phonon state in a wide wavevector range

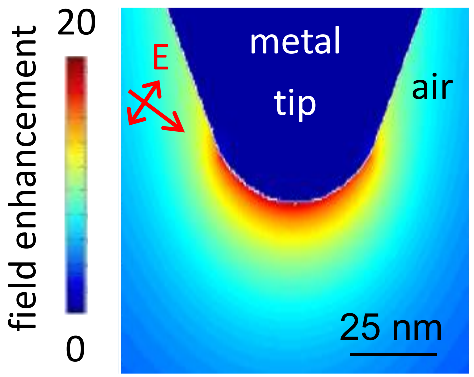
Detecting polariton with scattering scanning near-field optical microscopy (s-SNOM)

Focused laser beam illuminates AFM tip

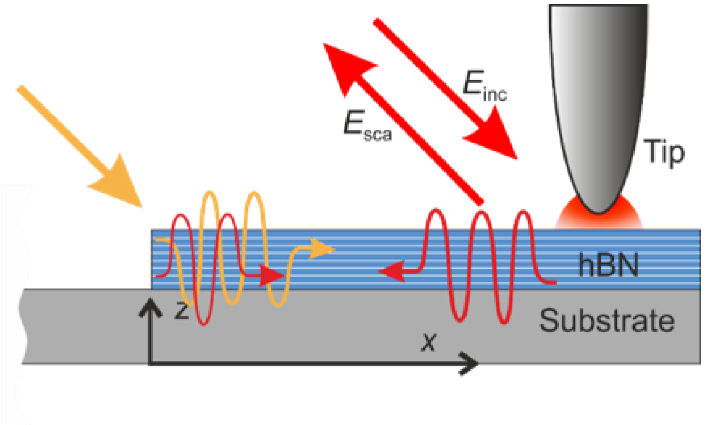
Detection of back scattered light



Tip creates nano-focus

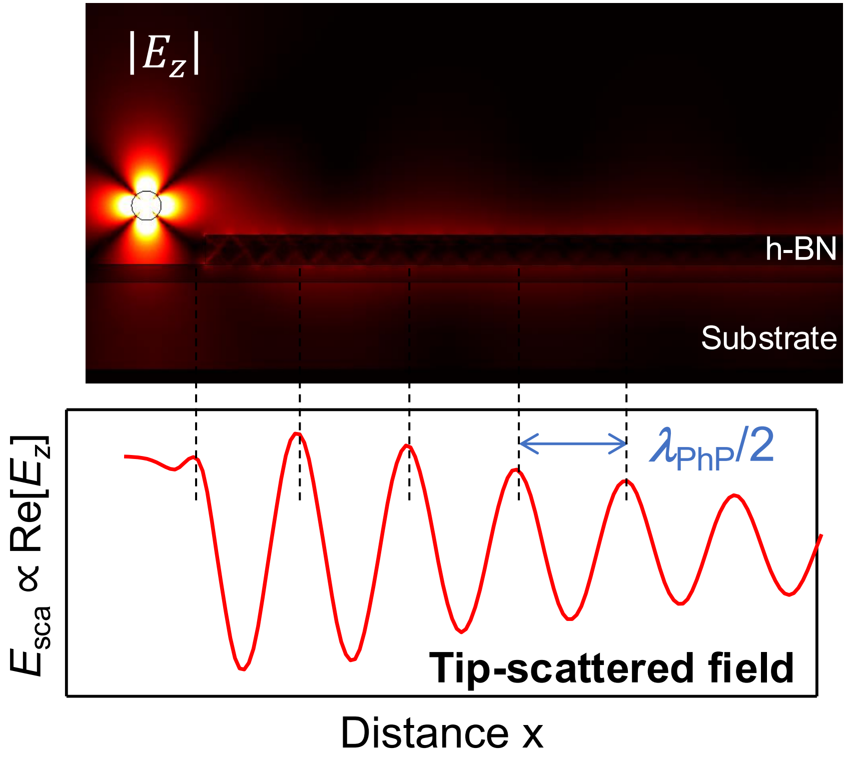


λ ≈ 10 µm



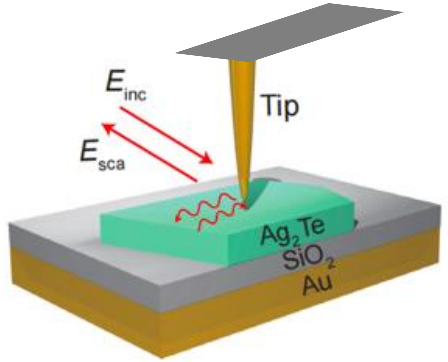
Polariton interferometry for imaging polaritons

Point dipole is mimicking the tip



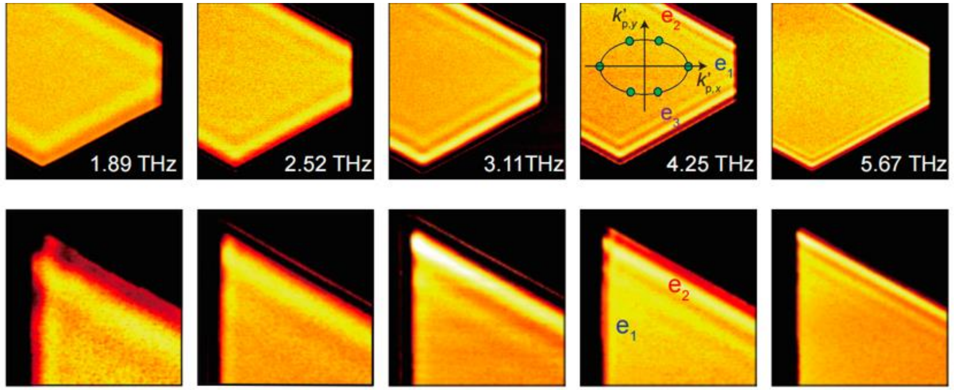
A. Bylinkin et al., *Nature Photon.* **15**, 197 (2021)

Example of polariton interferometry in anisotropic material

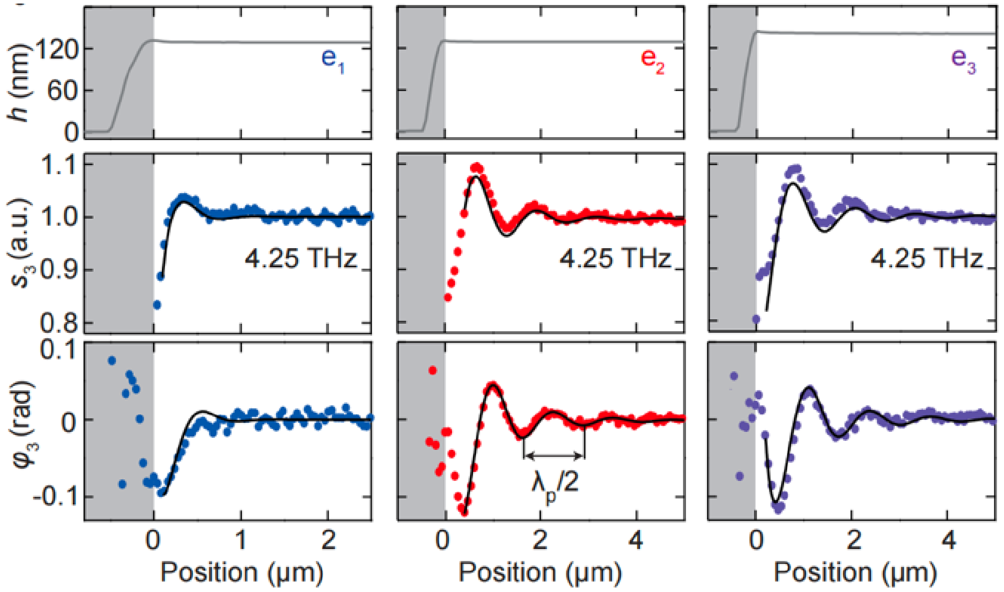


Surface plasmon polaritons in Ag2Te

THz near-field amplitude



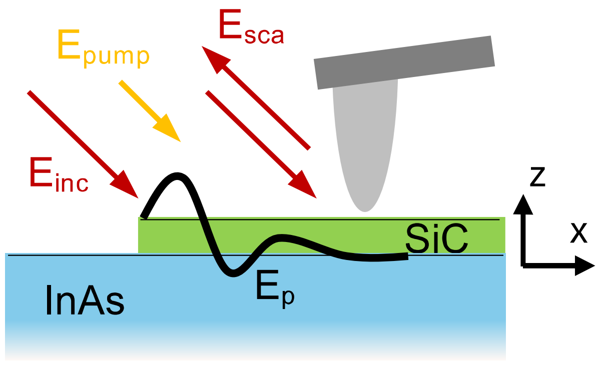
THz amplitude and phase line profiles



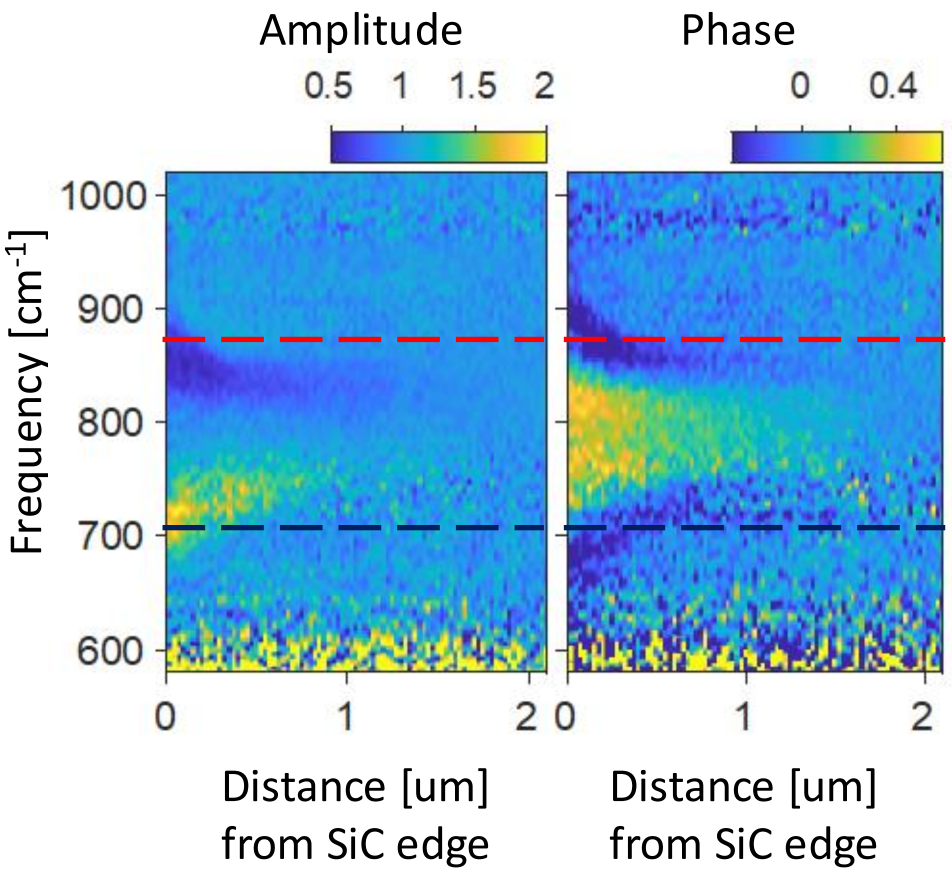
Few oscillation due to small group velocity and high damping

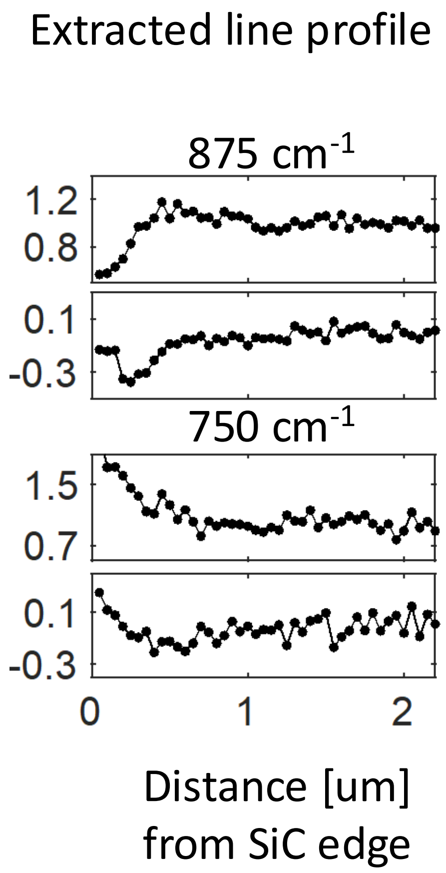
Chen, S. *Nat. Mater.* **22**, 860–866 (2023).

Polariton interferometry of InAs/SiC sample under photodoping



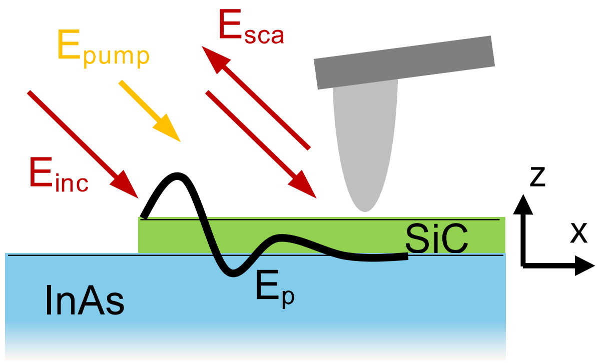
SiC edge launch polariton which is scattered by the AFM tip





Photoexcited InAs
From $n=10^{17}$ cm^{-3}
to $n=2x10^{18}$ cm^{-3}

Line profiles can be fitted by a decaying plane wave, yielding polariton wavevector



SiC edge launch polariton which is scattered by the AFM tip

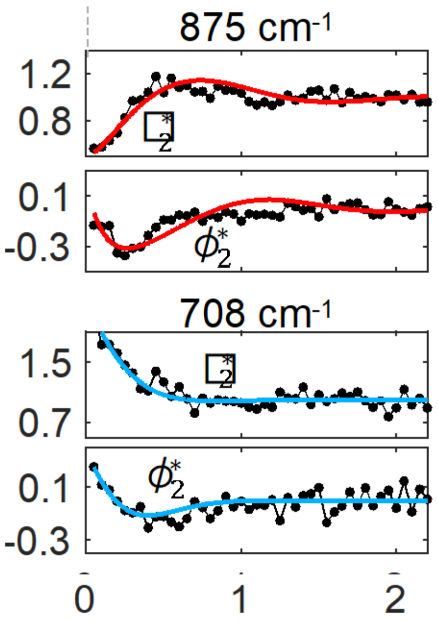
Photoexcited InAs
From n=10^{17} cm^{-3}
to n=$2x10^{18}$ cm^{-3}

We fit the polariton field with

$$E_p = E_0 e^{ikx} e^{-\gamma x}$$

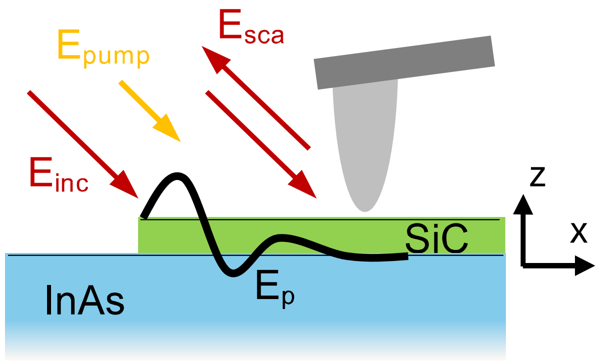
wavevector k

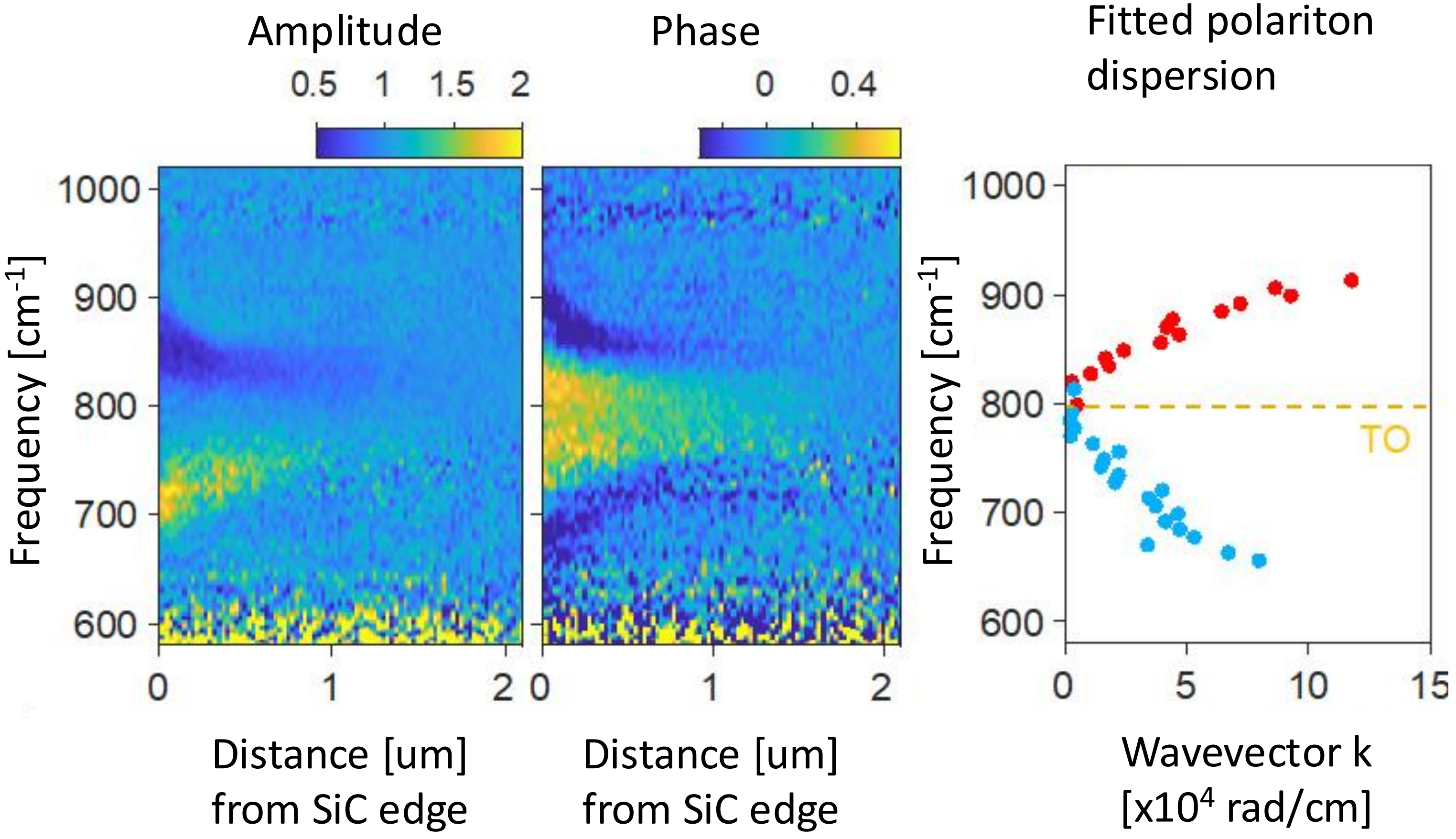
Extracted line profile



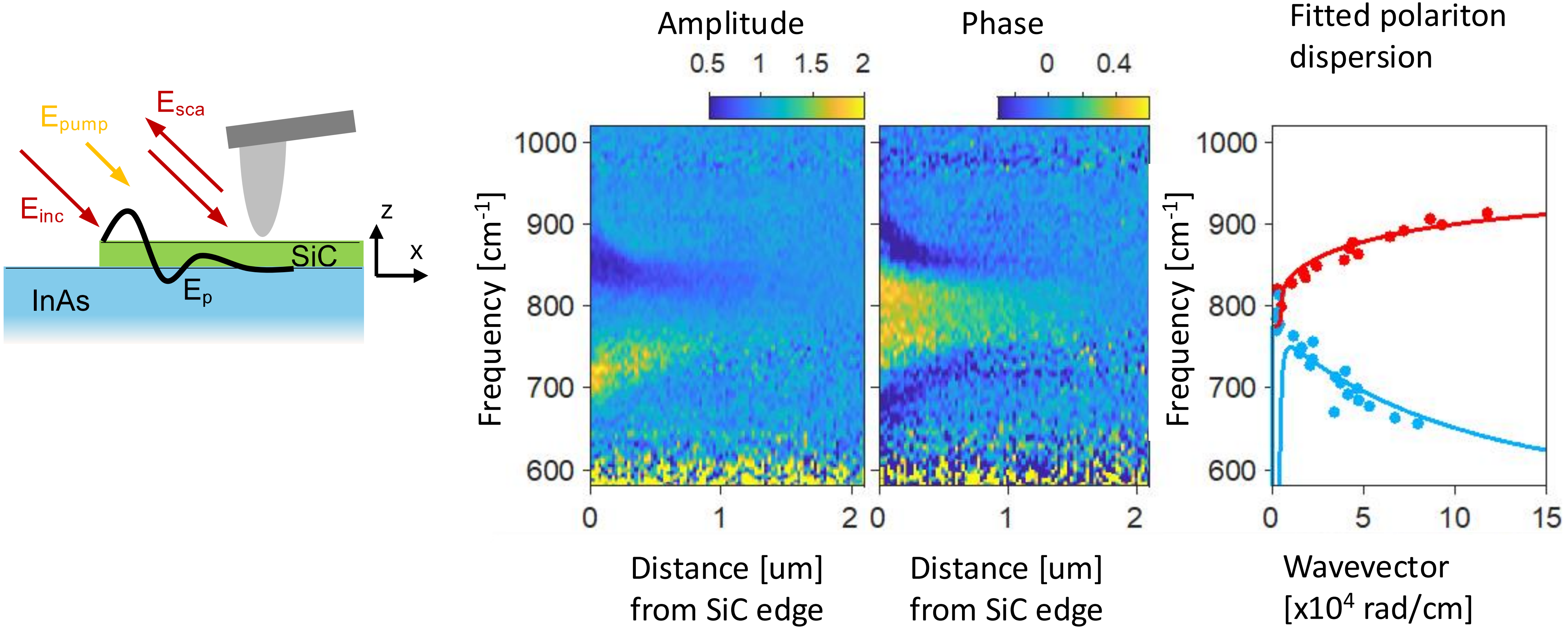
Distance [um] from SiC edge

Fitting for all frequencies yields dispersion





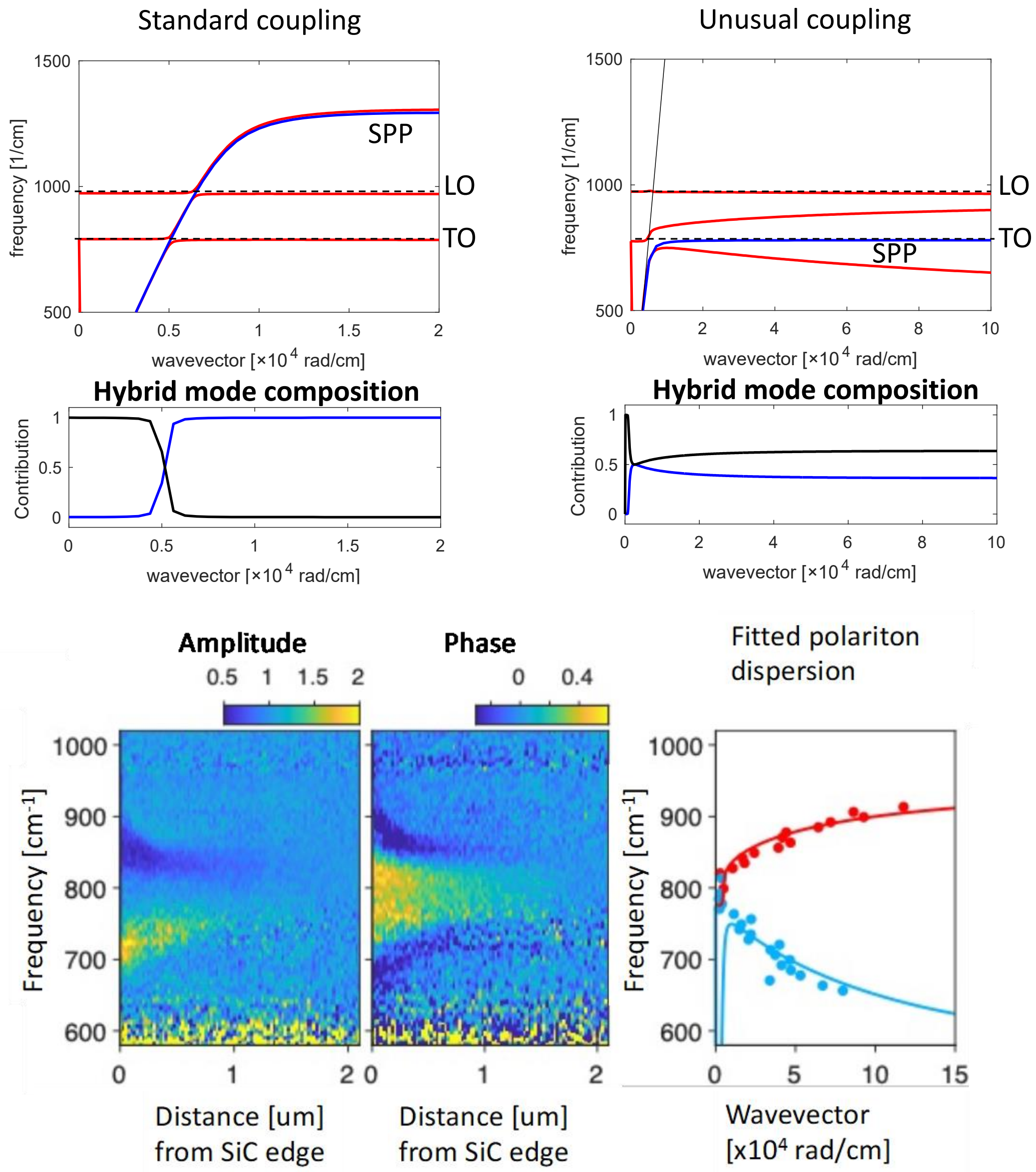
Exp. dispersion matches well the one obtaine from transfer matrix calcs



mixed polariton–phonon state in a wide wavevector range experimentally confirmed

Conclusion

- We studied the coupling between surface plasmon polariton in InAs with TO phonons in SiC.
- We predicted an unusual coupling when we match SPP dispersion with TO.
- We confirmed the predictions by measuring the polariton dispersions by polariton interferometry with s-SNOM.



Acknowledgements

CIC nanoGUNE
MEMBER OF BASQUE RESEARCH & TECHNOLOGY ALLIANCE

Nanooptics group

CFM Centro de Física de Materiales
Materialen Fisika Zentroa
Materials Physics Center

Xabier Arrieta
Ruben Esteban

DIPC

Javier Aizpurua

Fundings:

MINISTERIO DE CIENCIA, INNOVACIÓN Y UNIVERSIDADES
Financiado por la Unión Europea NextGenerationEU
Plan de Recuperación, Transformación y Resiliencia
AGENCIA ESTATAL DE INVESTIGACIÓN

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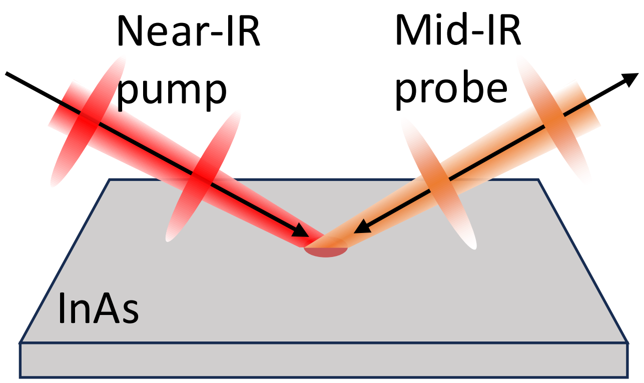
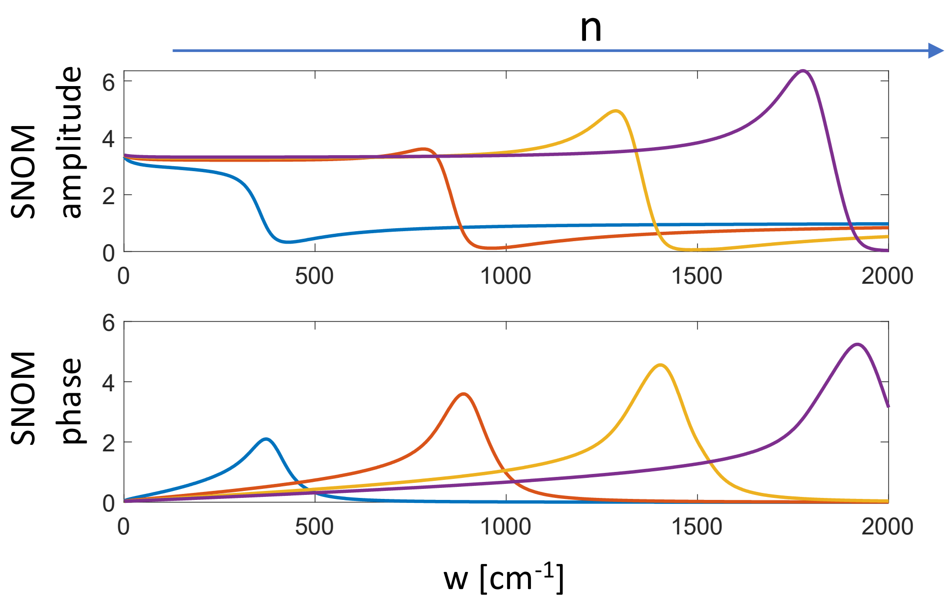
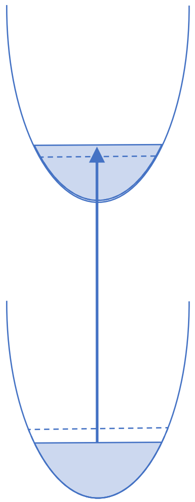
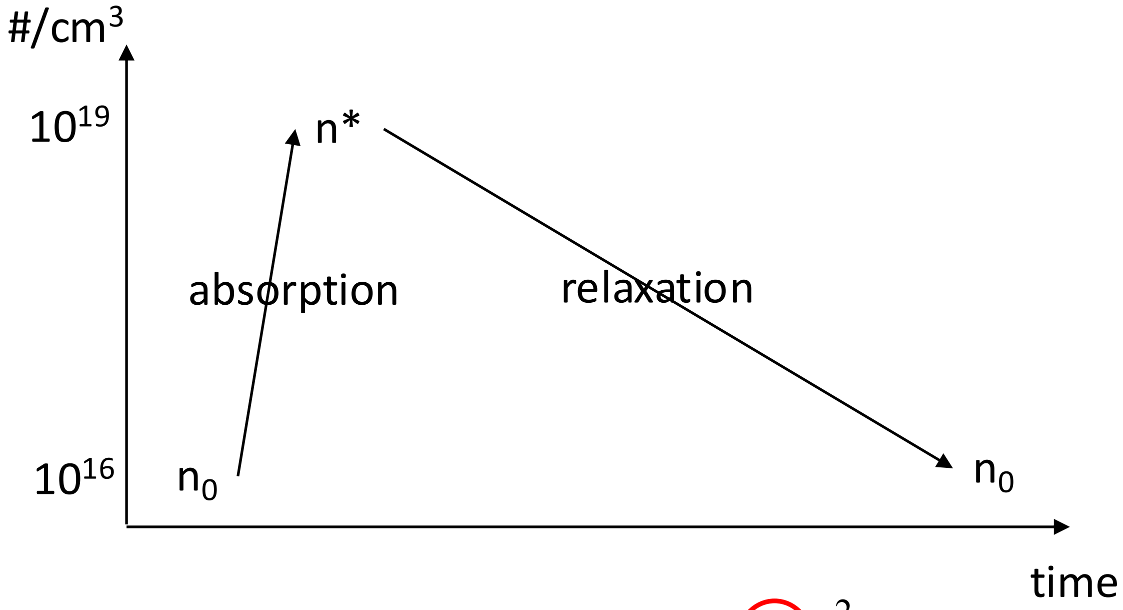
Thank you for the attention

Pump-probe nanospectroscopy





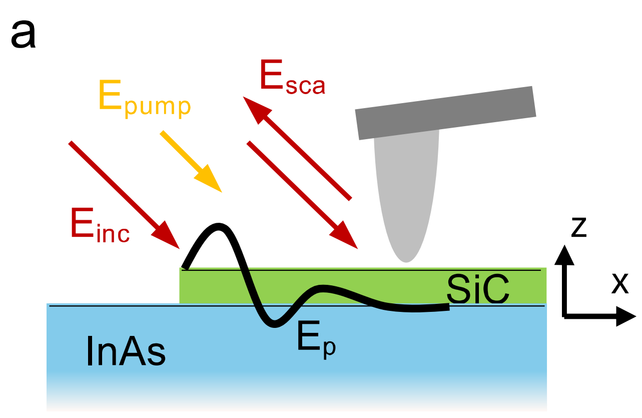


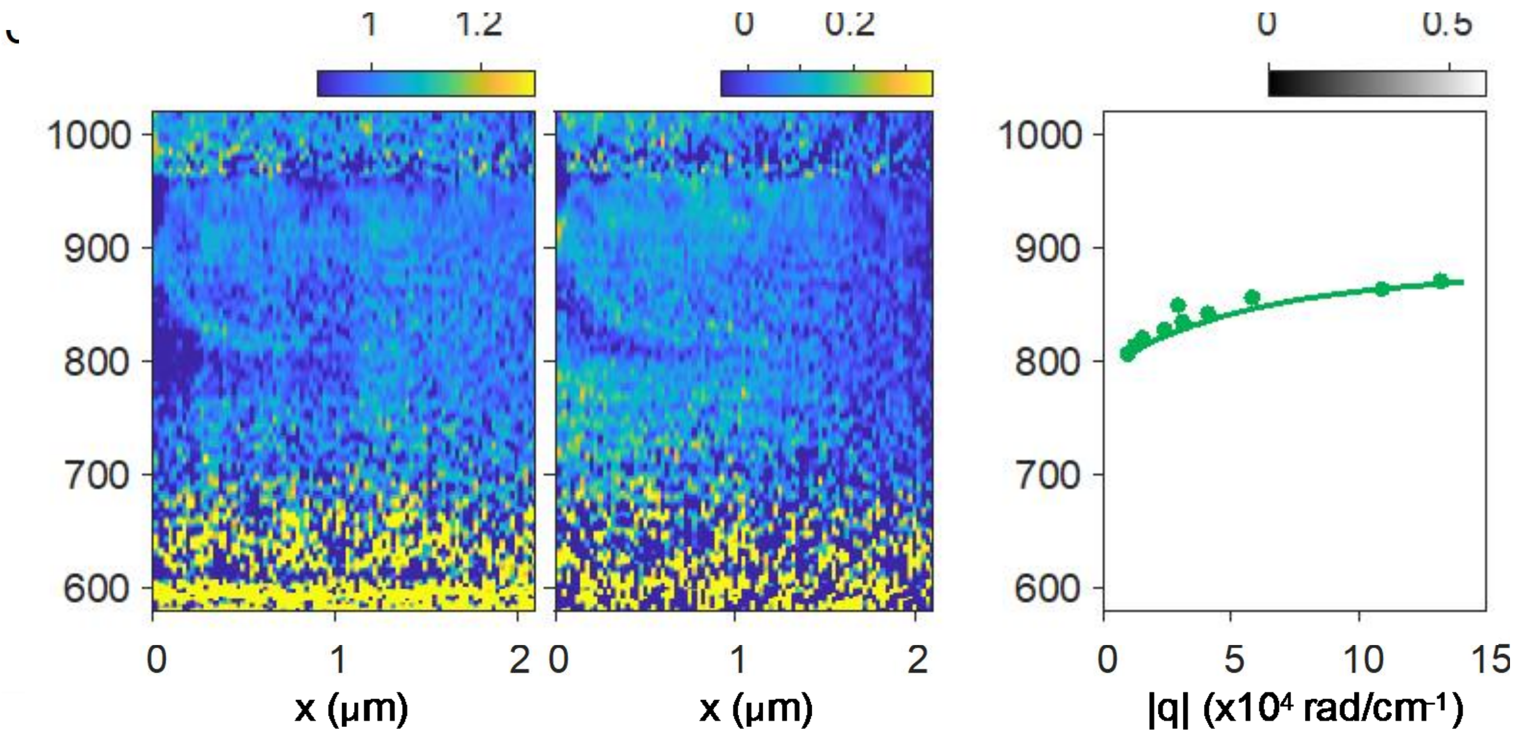


$$\varepsilon(\omega) = \varepsilon_\infty \left(1 - \frac{\omega_P^2}{\omega^2 + i\gamma\omega}\right) \qquad \omega_P^2 = \frac{n\, e^2}{\varepsilon_0 \varepsilon_\infty m^* m_0}$$

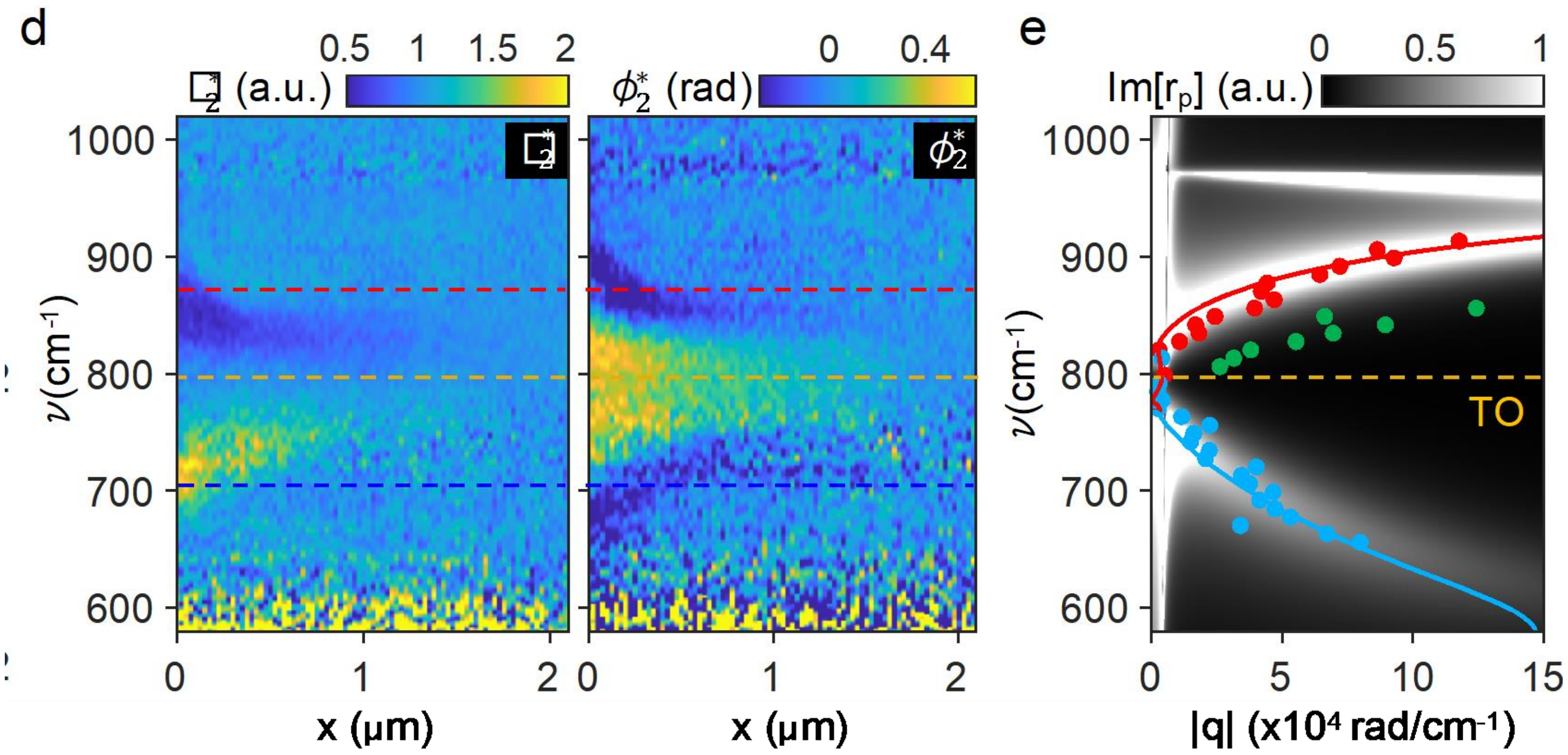
Pump-probe nanospectroscopy to photoexcite carriers in semiconductors and probe plasmon response

Polariton interferometry InAs/SiC

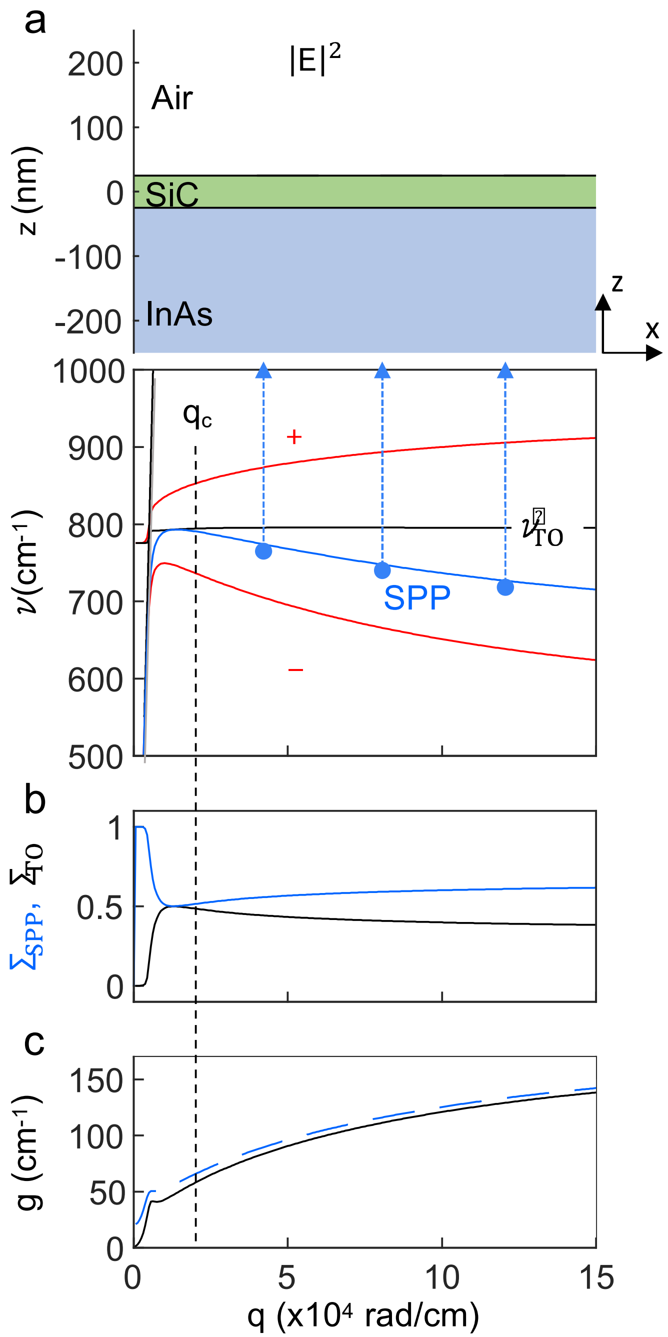




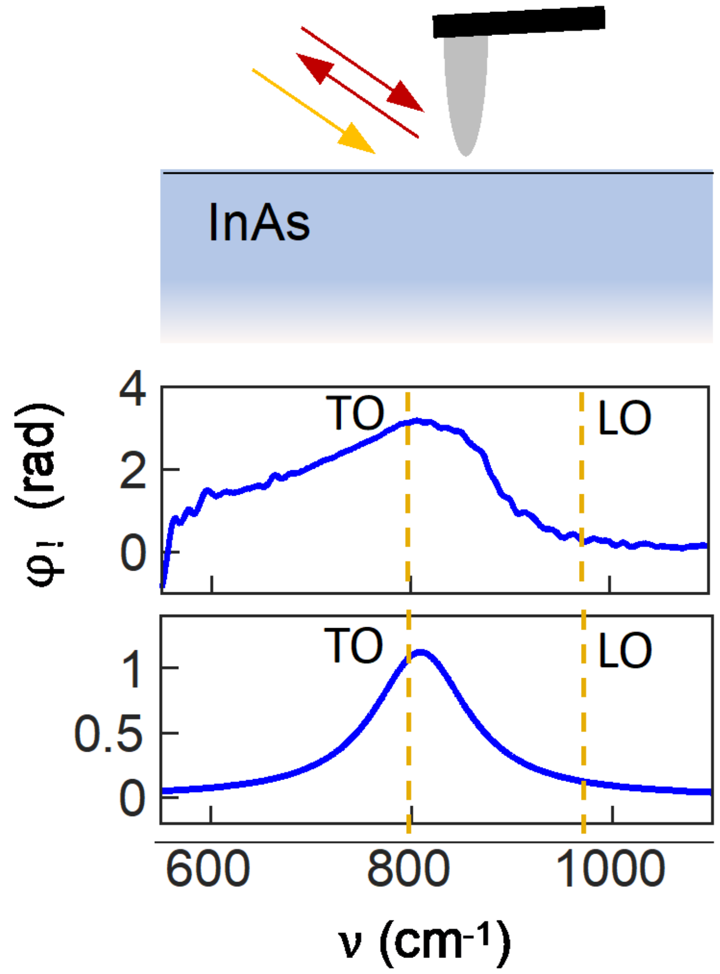
InAs
$n=10^{17}$ cm^{-3}



d
0.5 1 1.5 2
(a.u.)
0 0.4
ϕ_2^* (rad)
1000
900
800
700
600
ν(cm^{-1})
0 1 2
x (µm)
0 1 2
x (µm)
e
0 0.5 1
Im[r$_p$] (a.u.)
1000
900
800
700
600
ν(cm^{-1})
TO
0 5 10 15
|q| (x10^4 rad/cm^{-1})

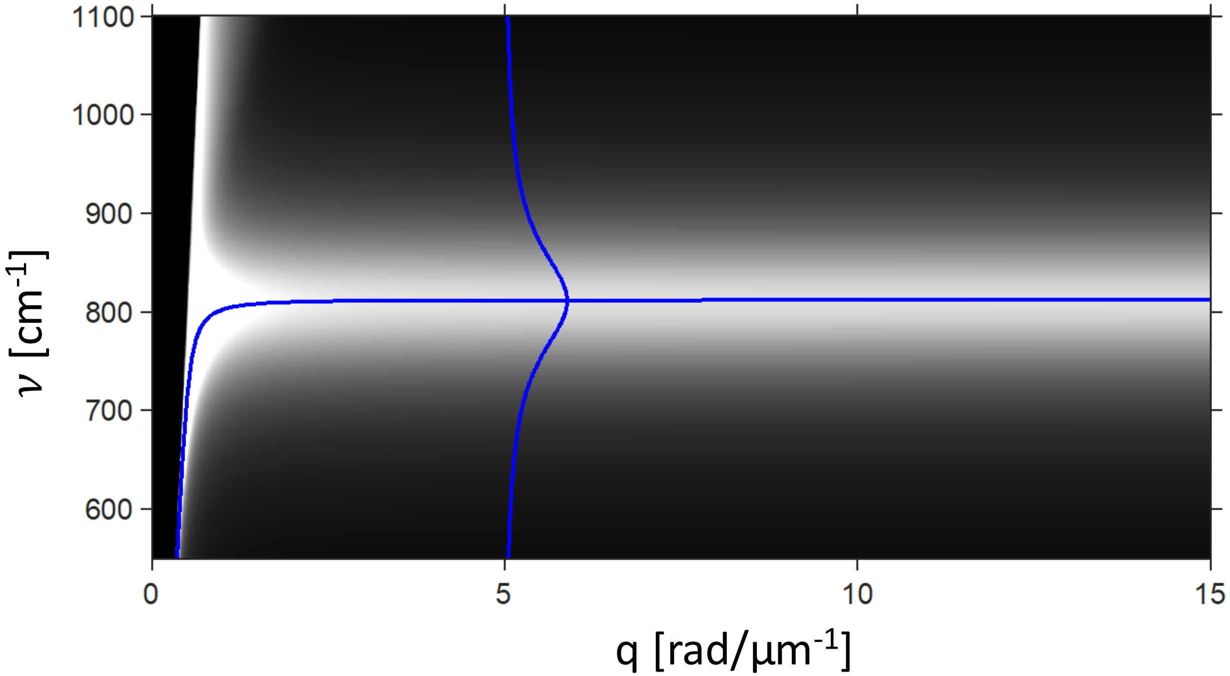

a
|E|^2
Air
SiC
InAs
z (nm)
200
100
0
-100
-200
z
x
1000
900
800
700
600
500
q_c
+
−
SPP
ν(cm^{-1})
b
1
0.5
0
Σ_{SPP}, Σ_{TO}
c
150
100
50
0
g (cm^{-1})
0
5
10
15
q (x10^4 rad/cm)

Pump-probe nanoFTIR of InAs



TM reflectivity as function of frequency $[\omega]$ and in-plane momentum $[q]$
Reflectivity poles -> mode dispersion

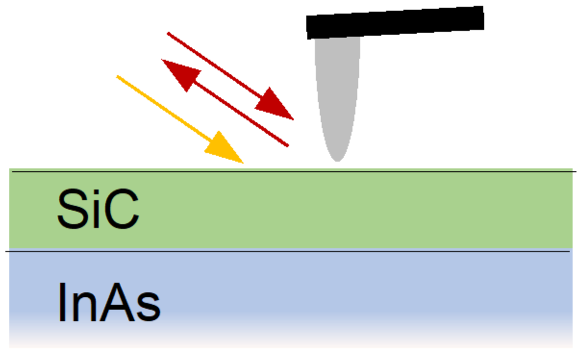
AFM tip $k \sim 1/R_{tip}$

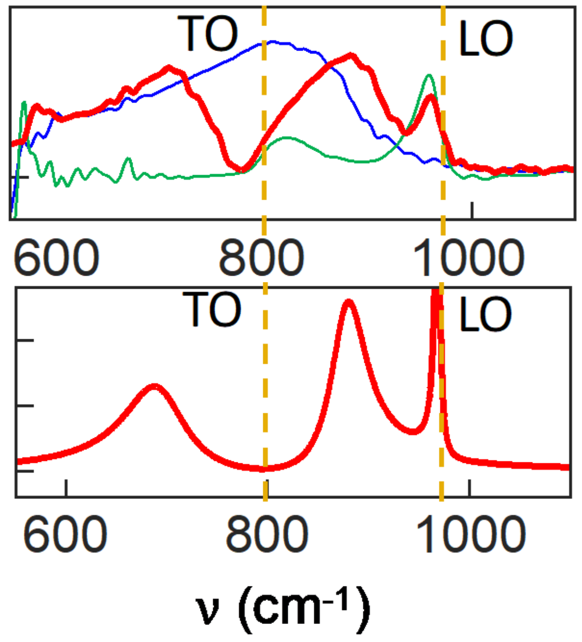


$\Delta t = 8.5$ ps

$\varepsilon_{\mathrm{InAs}}(\nu)$
$\nu_{\mathrm{p}} = 845\ \mathrm{cm}^{-1}$
$\gamma_{\mathrm{p}} = 125\ \mathrm{cm}^{-1}$

Pump-probe nanoFTIR of InAs/SiC





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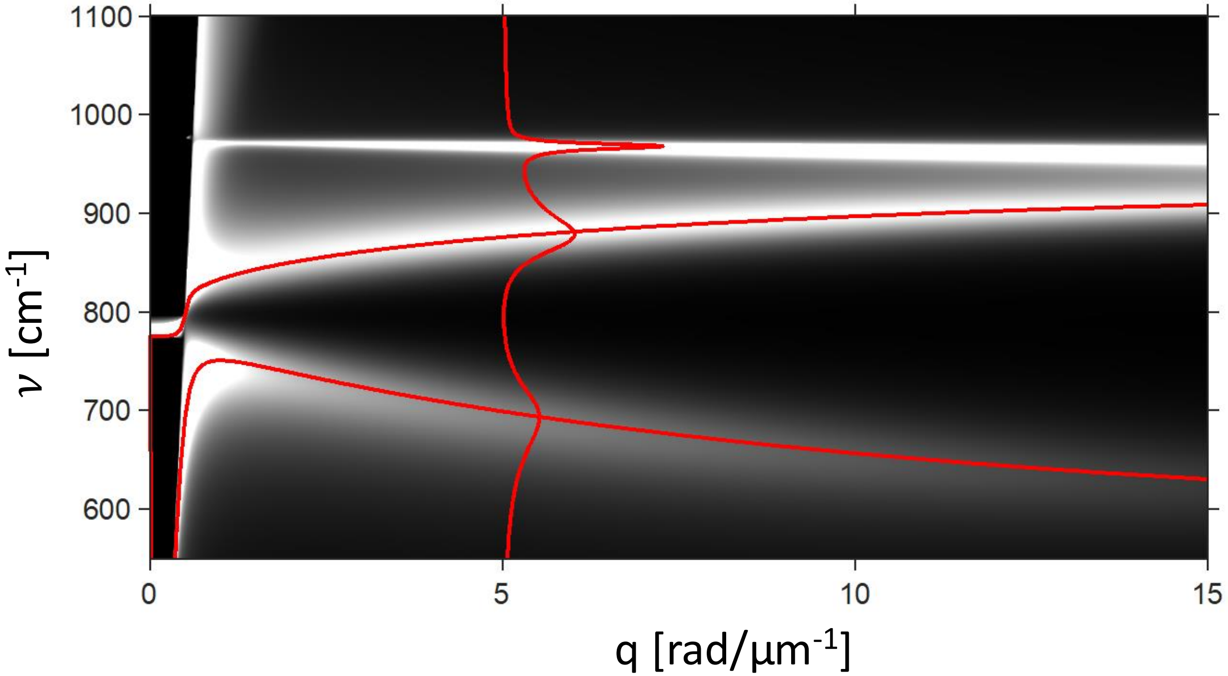

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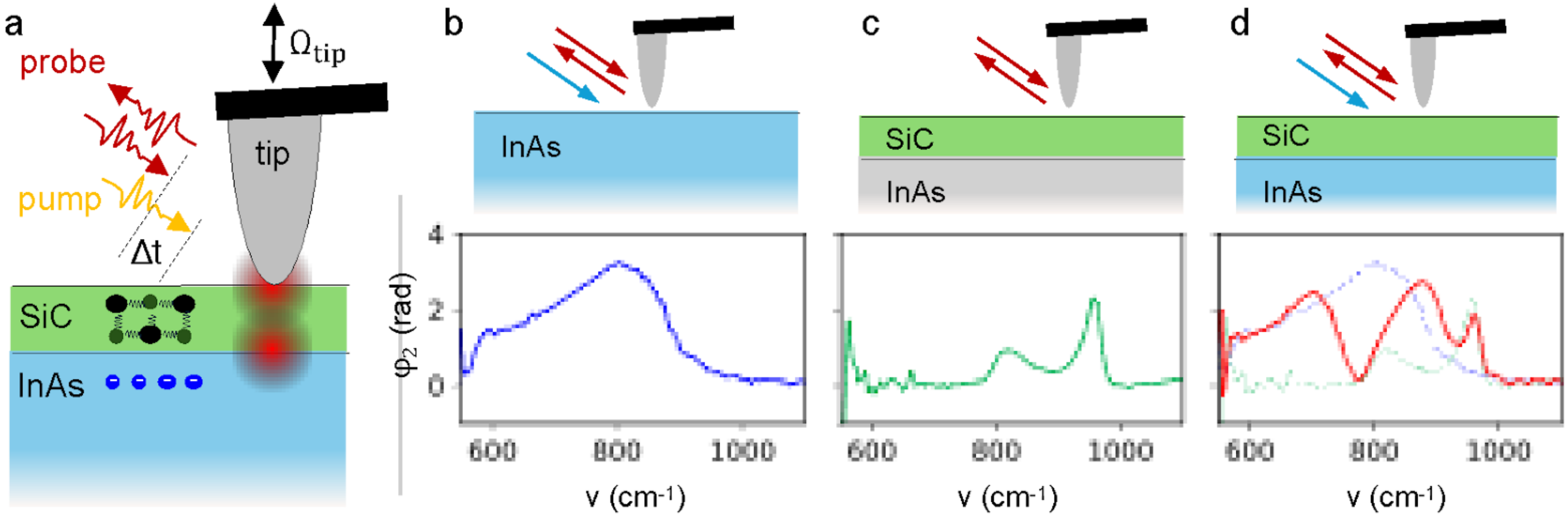

a
probe
pump
Δt
tip
Ω_{tip}
SiC
InAs
b
InAs
c
SiC
InAs
d
SiC
InAs
φ_2 (rad)
4
2
0
600
800
1000
ν (cm-1)
600
800
1000
ν (cm-1)
600
800
1000
ν (cm-1)