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Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering

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Technical Note

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14. ABSTRACT
Electromagnetic radiation is emitted by the vibrational and collective model of an opaque solid as the result of impulsive stimulated Raman scattering. Raman scattering of near-infrared femtosecond laser pulses produces coherent longitudinal optical phonon and plasmon oscillations in the semiconductor InSb. These oscillations radiate into free space at THz frequencies and are directly detected. The THz spectra exhibit features consistent with Raman selection rules including

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EXECUTIVE SUMMARY

The direct detection of THz radiation produced by stimulated Raman scattering of near infrared ultrafast laser light is reported. A frequency doubled Nd:YAG laser is used to pump a titanium doped sapphire crystal to produce an ultrafast laser beam. The laser emission from this source irradiates a semiconductor InSb sample, exciting optical phonon and plasmon oscillations which re-emit at ThZ frequencies. A bolometer is used to detect the THZ radiation as a function of the azimuthal angle of the InSb crystal. The observation of an azimuthal dependence of the THz intensity is indicative of a coherent process and confirms the generation of stimulated Raman scattering.

Excitation of coherent phonons and plasmons in InSb by impulsive Raman scattering

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Introductory vg giving title of talk. All the information provided has previously been cleared in AFRL/DE 23-1 (2 Jan 2003) entitled "Terahertz (THz) Radiation Generation, Physics and Applications" and AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering". The second cleared abstract and write-up was a consolidation of the AFRL/DE 23-1 cleared presentation. Although all of the information has been cleared previously in other presentations, this presentation is being processed for clearance since there are different formats for presentation clarity. All of work is 6.1 research funded by AFOSR

Measure THz spectra of bulk InSb

Main Results:

Coherent phonons and plasmons are selectively excited by ultrafast pulse via Raman scattering

Coherent longitudinal modes radiate into free space

(100) surface: Raman mechanisms interfere

(111) surface: Raman excitation without interference

List of main results being presented. vg shows how THz radiation produced – previously described in paper entitled "Emission of Terahertz Radiation from Coupled Plasmon-Phonon Modes from InAs", AFRL-DE 21-681, 20 Dec 2001.

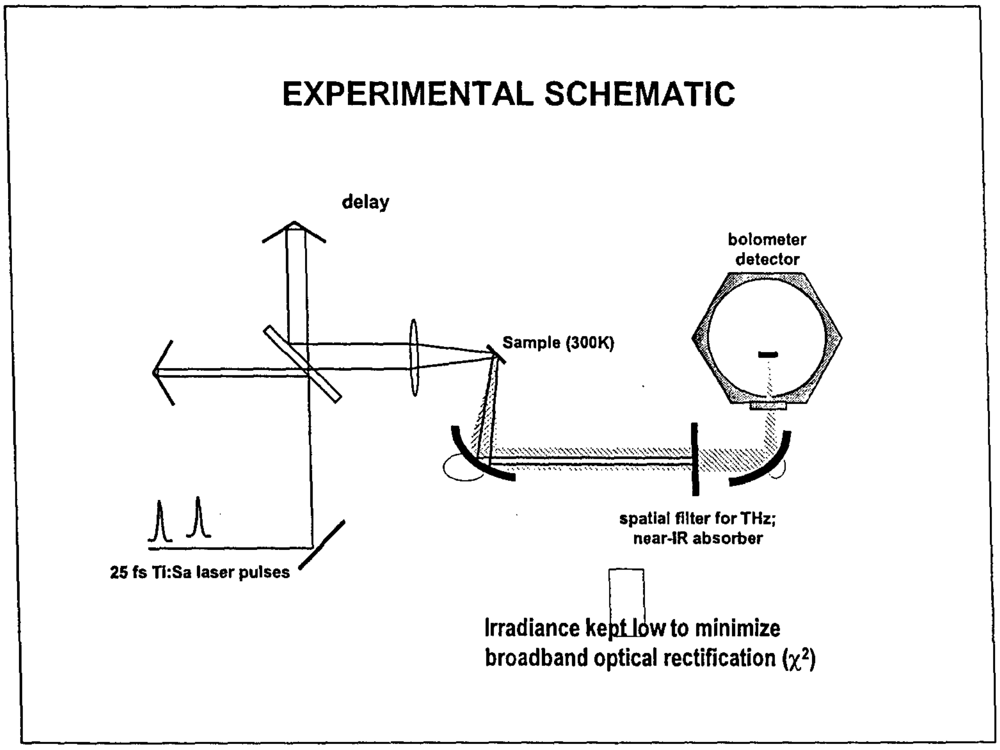
THz detection techniques

Gated photonducting antennas
Severe response rolloff > 3 THz

Electro-optic sampling with ZnTe
Reststrahl absorption/dispersion 5-6 THz

Interferometeric autocorrelation
Broadband; No phase information
(Bonvalet et al, APL, 1995; Kersting et al, PRL, 1997)

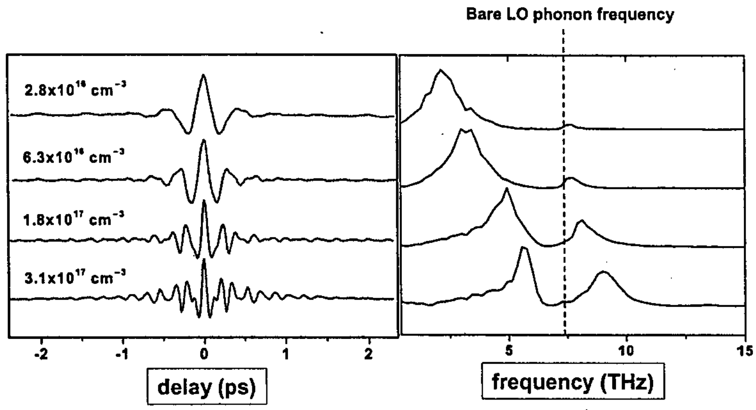
List the types of Terahertz detection techniques used throughout world. These approaches we have reported previously plus extensively described throughout world in technical archieval journals



Experimental method used to measure THz radiation. Shown previously as cleared presentations. Same as vg #7 and 18 in in AFRL/DE 23-1 (2 Jan 2003) entitled "Terahertz (THz) Radiation Generation, Physics and Applications"

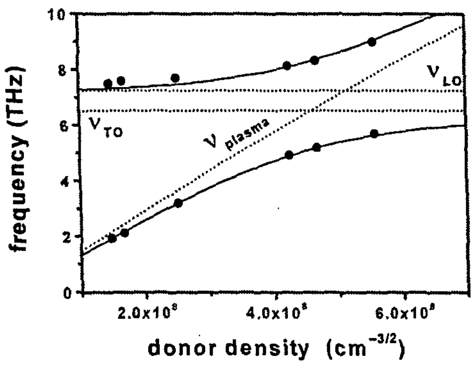
Experiments with bulk n-InAs

(Hasselbeck et al, Phys. Rev. B, 2002)



Same data as show in vg # 20 of cleared presentation in AFRL/DE 23-1 (2 Jan 2003) entitled "Terahertz (THz) Radiation Generation, Physics and Applications"

n-InAs: Results



Dispersion of coupled plasmon-phonon modes is exactly predicted

Plasma frequency unaffected by photocarriers ($N > 10^{18}$ cm^{-3})

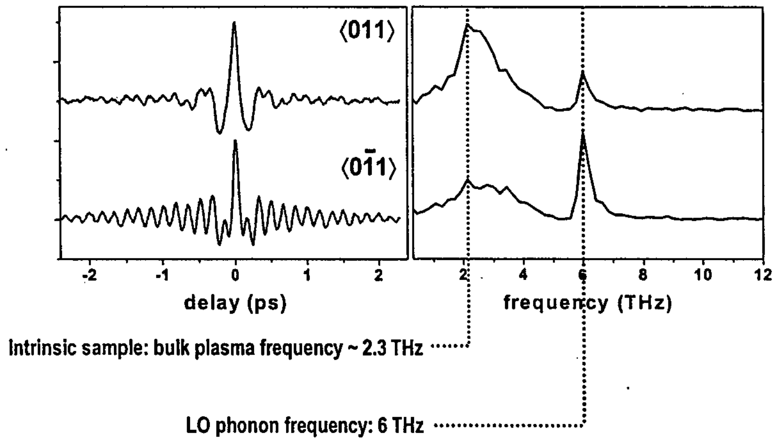
Independent of sample growth direction: (100) or (111)

Independent of azimuthal orientation of laser polarization

Coherent oscillations started by ultrafast field transient:
- ***surface field screening***
- ***photo-Dember effect***

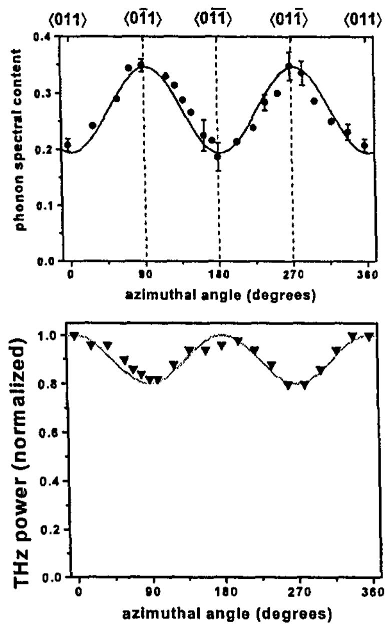
Same data as show in vg # 22 of cleared presentation in AFRL/DE 23-1 (2 Jan 2003) entitled "Terahertz (THz) Radiation Generation, Physics and Applications"

Strong azimuthal effect observed with (100) InSb



Vg shows variation of effect of THz radiation in InSb. Described in cleared presentation cleared in AFRL/DE 23-1 (2 Jan 2003) entitled "Terahertz (THz) Radiation Generation, Physics and Applications" and AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering"

Phonon content = $\frac{\int phonon\ spectrum}{\int total\ spectrum}$



Spectra cannot be explained by surface rectification

Interference of Raman excitation mechanisms

Vg shows THz emission spectra from InSb and is exact Fig.2 of cleared document AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering"

Raman excitation of longitudinal modes in opaque semiconductors

Deformation potential (forbidden) → drives exclusively phonons

Charge density fluctuations (allowed) → drive exclusively plasmons

Electro-optic mechanism (allowed) → drives both

$$\left.\begin{array}{l} h\nu_{laser} = 1.6\ eV \\ E_g + \Delta \approx 0.98\ eV \end{array}\right\}$$

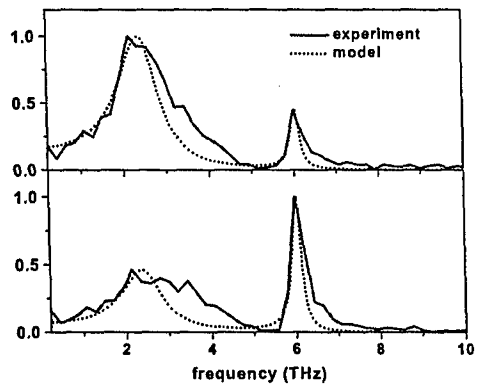
Elements of Raman tensor are complex
Interference is possible

Vg shows summary of two previous vg and has been cleared previously in AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering"

Spontaneous Raman scattering from (100) GaAs *(Chen, Phys. Rev. B, 1983)*

Interference of allowed and forbidden scattering mechanisms

Near resonance with $E_g + \Delta$ (split-off valence band – conduction band)



Chen model explains
THz spectra for (100) InSb

Vg shows THz emission spectra from InSb and is exact Fig.1 of cleared document AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering"

(111) InSb

- No Raman interference expected → no azimuthal dependence
- Optical rectification ~ 7x greater than (100) surface
  (Gu et al, J. Appl. Phys., 2002)
- Keep excitation conditions identical to (100) experiments

Summary of results which provided in this presentation and previously cleared in AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering"

(111) InSb : Normalized emission spectra

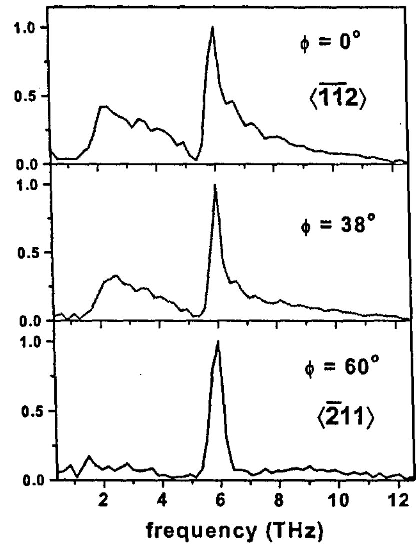
Broad background radiation from optical rectification [Difference frequency mixing of ultrafast pulse]

Three-fold azimuthal symmetry

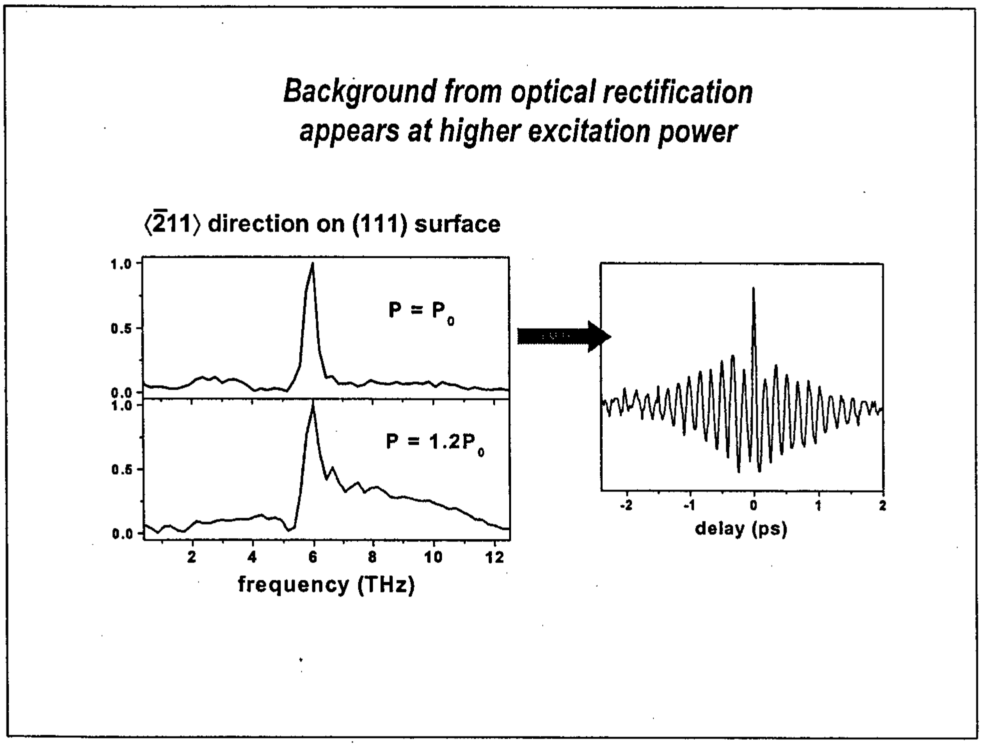
Null at TO phonon frequency (5.6 THz): phonon–polariton absorption

Background disppears when pump light polarized along <-211> direction

Emission entirely from coherent LO phonons!



Variation of results presented in this presentation (vg # 10) and cleared in document AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering"



Expansion of results presented in this presentation (vg # 10 and 12) showing effect of optical rectification and cleared in document AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering". Also, was cleared in AFRL/DE 23-1 (2 Jan 2003) entitled "Terahertz (THz) Radiation Generation, Physics and Applications"

(111) InSb: Radiation power

Bolometer detects:

- **Both s- and p-polarized optical rectification (OR) signals**
- **Background Raman signal (p-polarized)**

Azimuthal power dependence – Optical rectification *(Gu et al, J. Appl. Phys, 2003):*

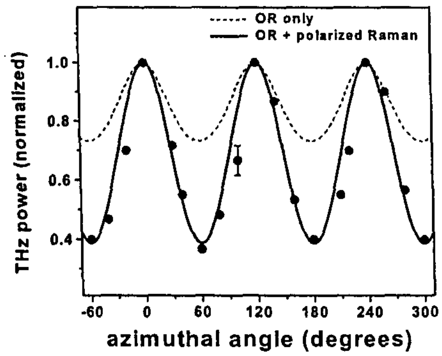
$$P_{THz} \propto [0.741\cos(3\theta) - 0.057 + R]^2 + 0.534\sin^2(3\theta)$$

***R:* constant Raman background from coherent phonons**

Fit data with *R* = 0.23

Spontaneous Raman scattering at 920 nm (cw excitation):

→ No azimuthal dependence
→ Constant Raman background



Expansion of summary of vg 12 in this presentation and previously cleared in AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering"

SUMMARY

- Low level excitation of InSb produces radiation by coherent phonons and plasmons
- Previously observed temperature dependence explained by shifting plasma frequency *(Hu et al, 1990; Howells et al, 1994; Kono et al, 2000; Gu et al, 2002)*
- No evidence for photo-Dember current in the spectra
- Coherent oscillations excited by impulsive Raman scattering
- Previously observed p-polarized background radiation from (111) InSb consistent with Raman selection rules *(Howells et al, 1995)*

Raman spectroscopy in the THz domain: direct measurement of the radiating mode!

Summary of presentation which was cleared in document AFRL/DE 23-209 (22 May 2003) entitled "Excitation of Coherent Phonons and Plasmons in InSb by Impulsive Raman Scattering"

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