ALS User's Meeting, Berkeley, CA
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Ultrafast X-ray Diffraction in Solids using K Radiation from Femtosecond Laser Generated Plasma Sources

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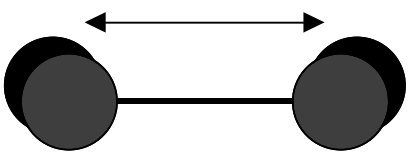
Universität Essen, Germany

Why ultrafast x-rays ?

Directly observe ultrafast atomic motion

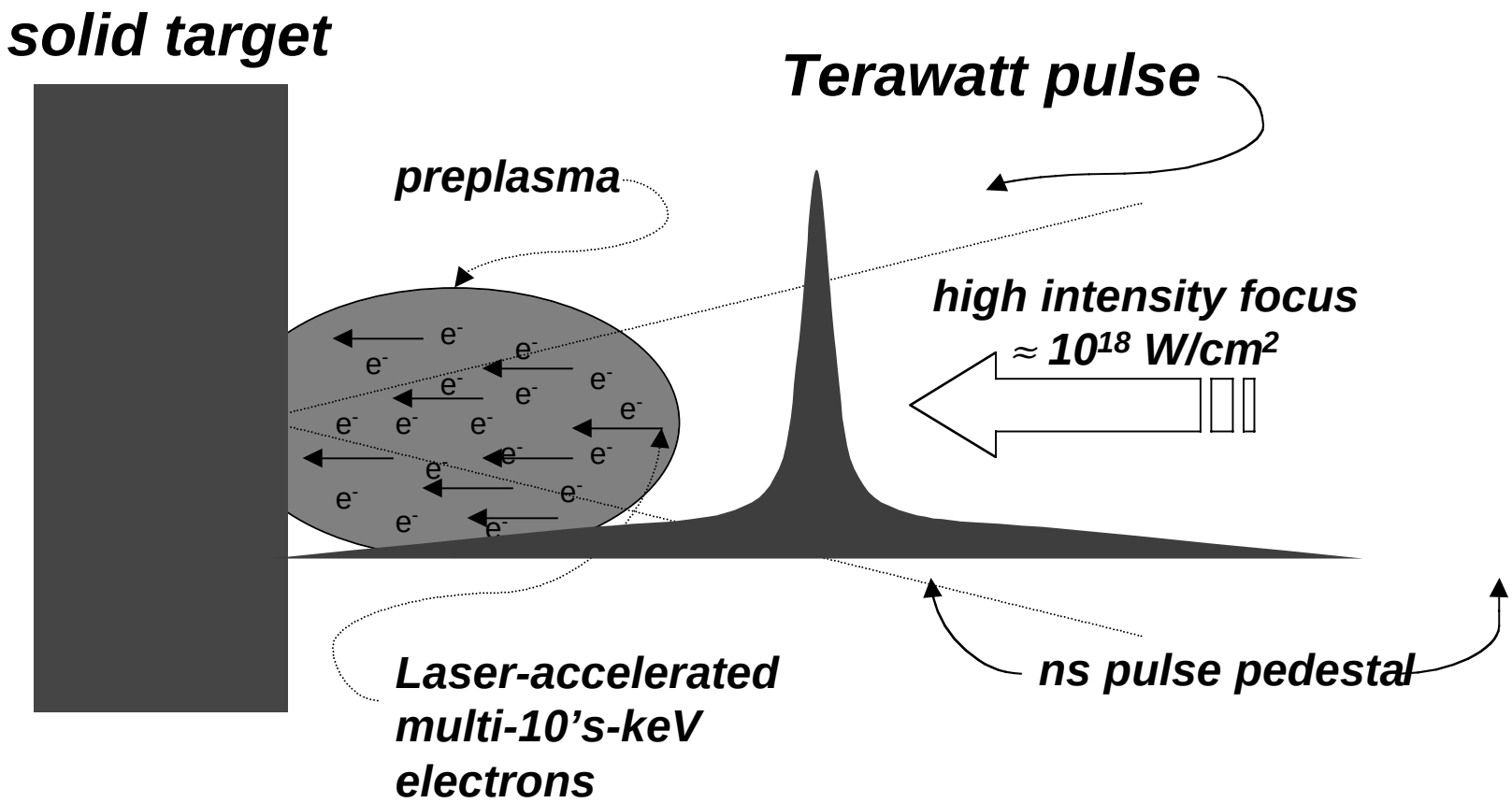
1 Å or ~10 keV



1) Short pulses (fs to ps)

2) Hard x-ray radiation

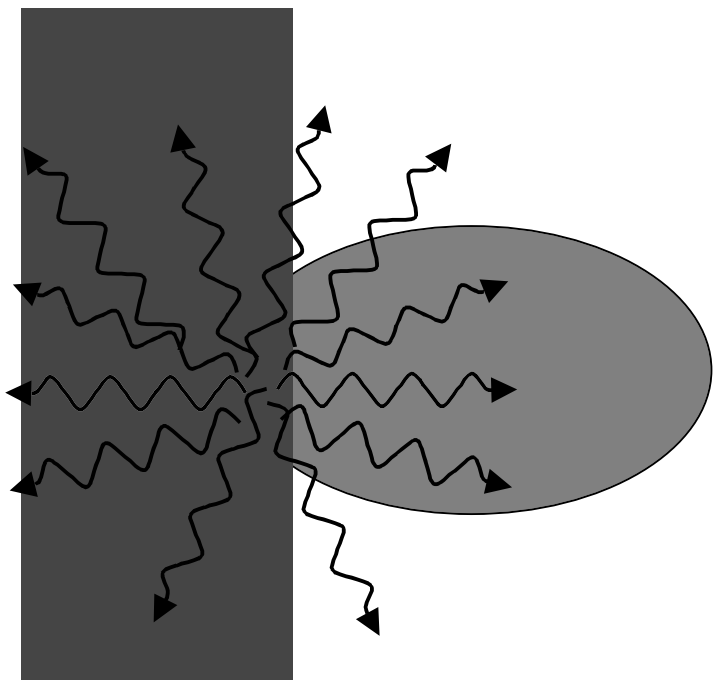
Laser-driven Electrons for Fast X-rays



J.D. Kmetec, et al., *Phys. Rev. Lett.* 68, 1527 (1990).

Laser-driven Electrons for Fast X-rays

solid target

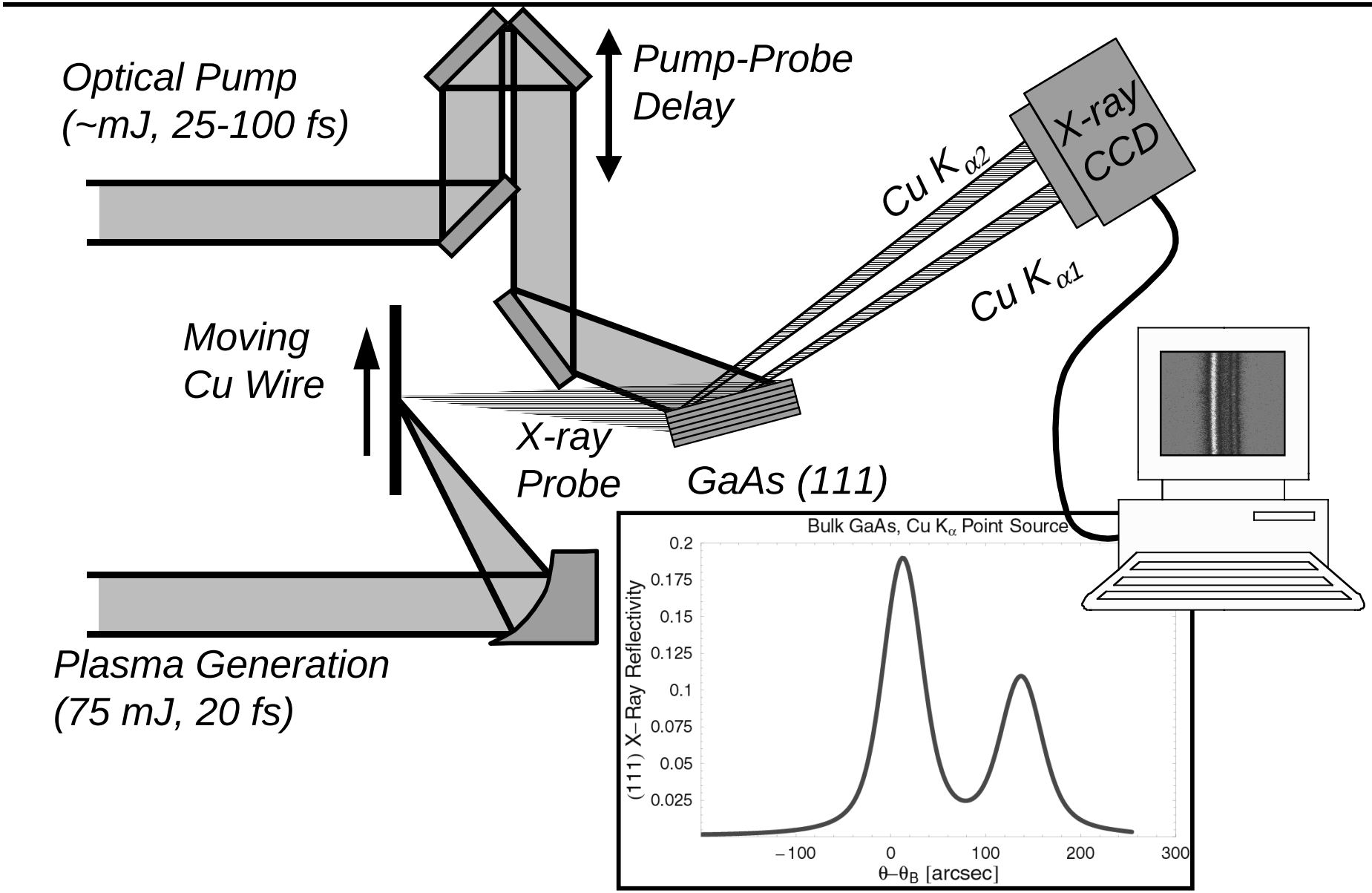


- **x-ray pulse duration: $\Delta t_{electron}$ and stopping time**
- **~ 200 fs at ENSTA**
- **Source size ~ laser spot**
- **10^9 photons / shot**

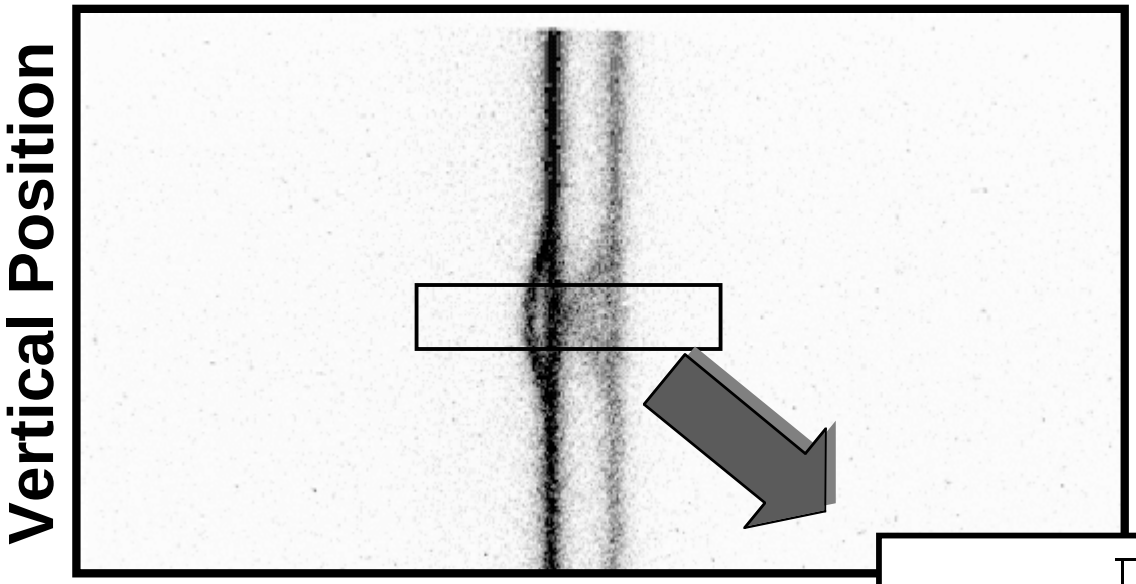
Bremsstrahlung and K_α radiation

J.D. Kmetec, et al., *Phys. Rev. Lett.* 68, 1527 (1990).

Ultrafast X-ray Diffraction



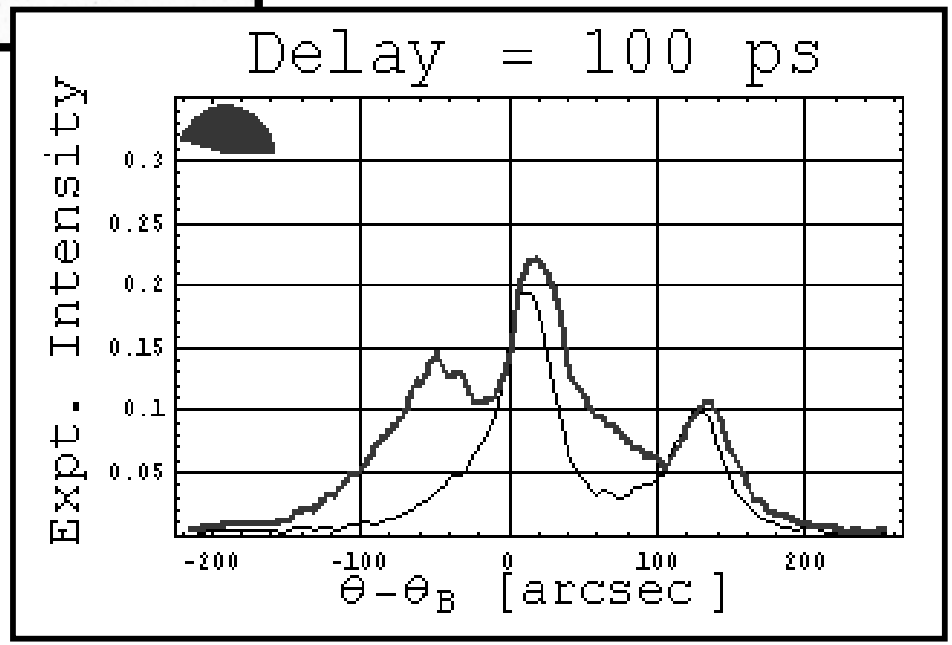
Bulk GaAs



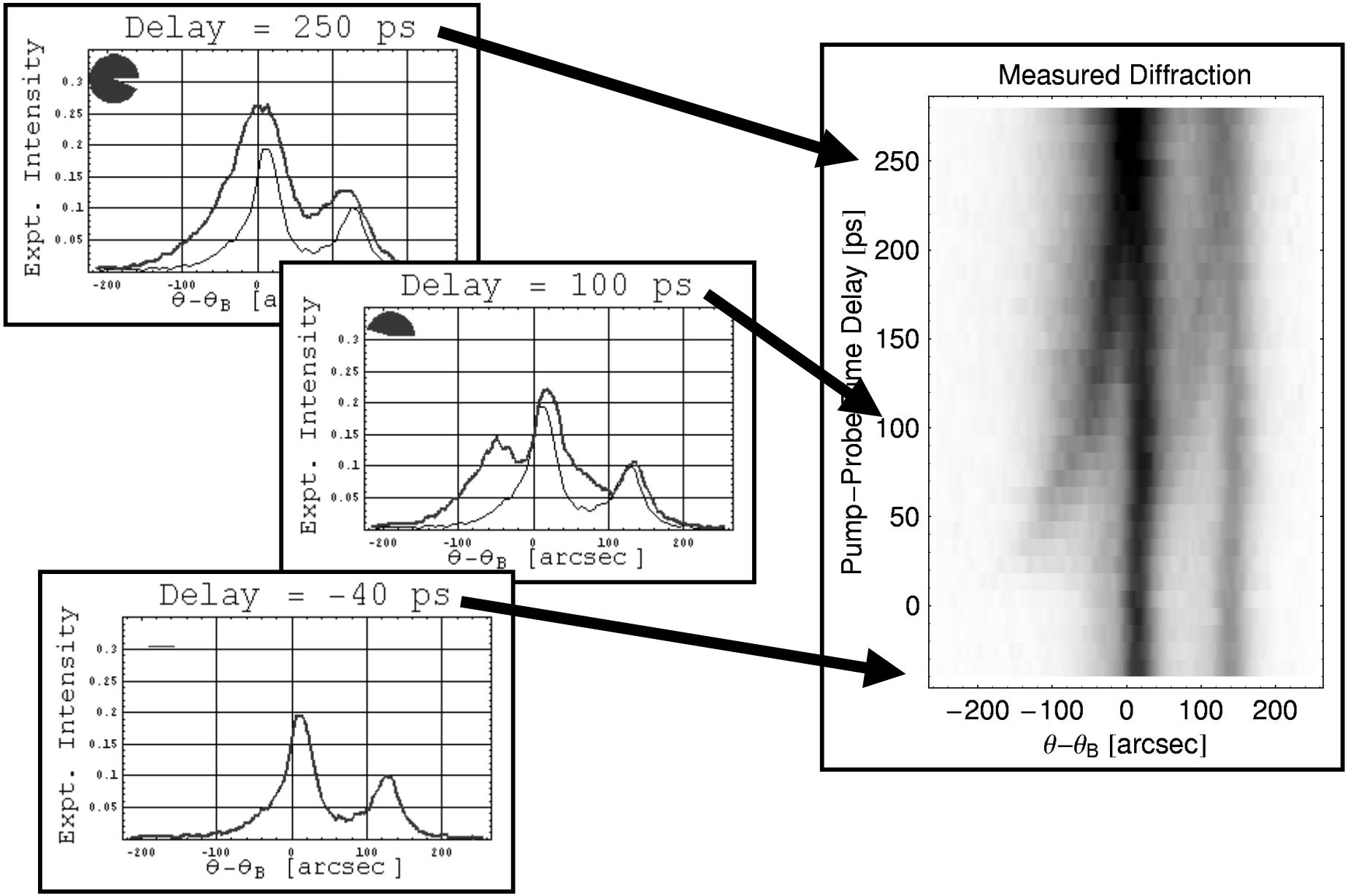
F = 50 mJ / cm²

τ = 30 fs

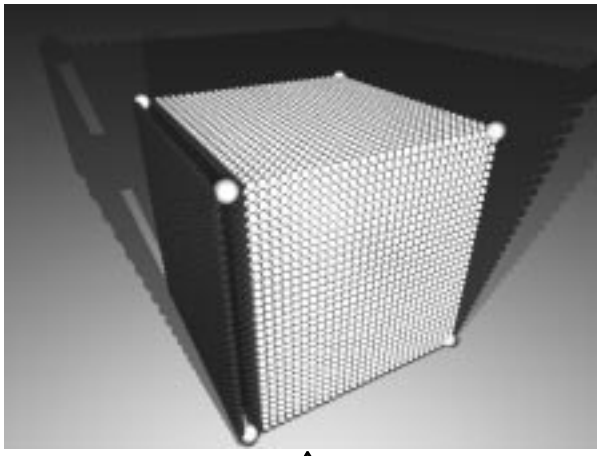
Integrated Lineouts

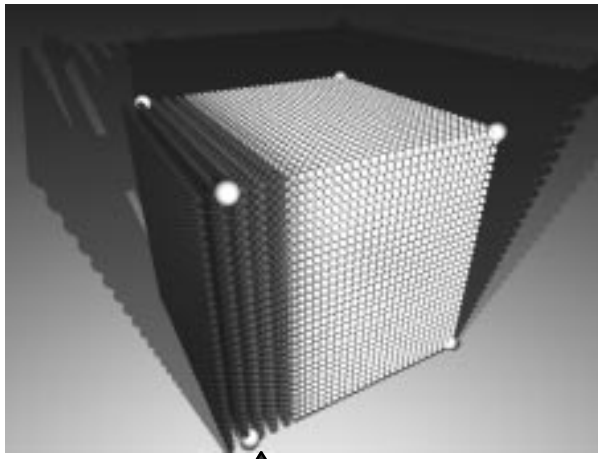


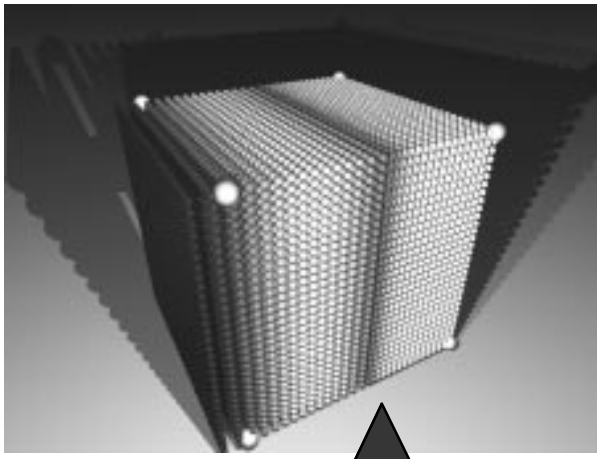
Ultrafast X-ray Diffraction:GaAs



Coherent Acoustic Phonons







Fast heating at constant Volume

Surface expansion

Newton's 3rd Law

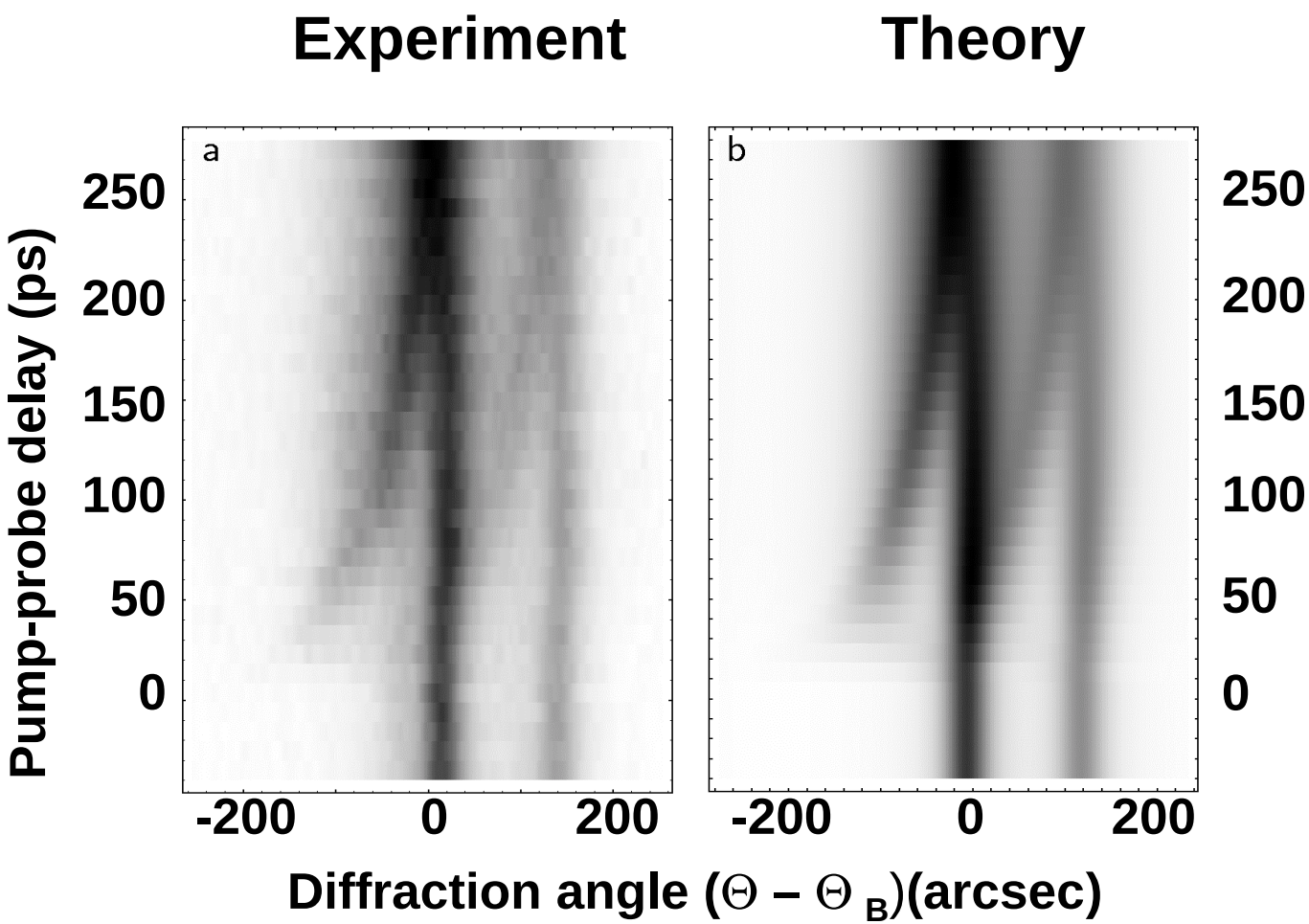
Stress

Strain

Acoustic Pulse

C. Thomsen et al. Phys Rev. B 34, 4129 (1986).

Gallium Arsenide

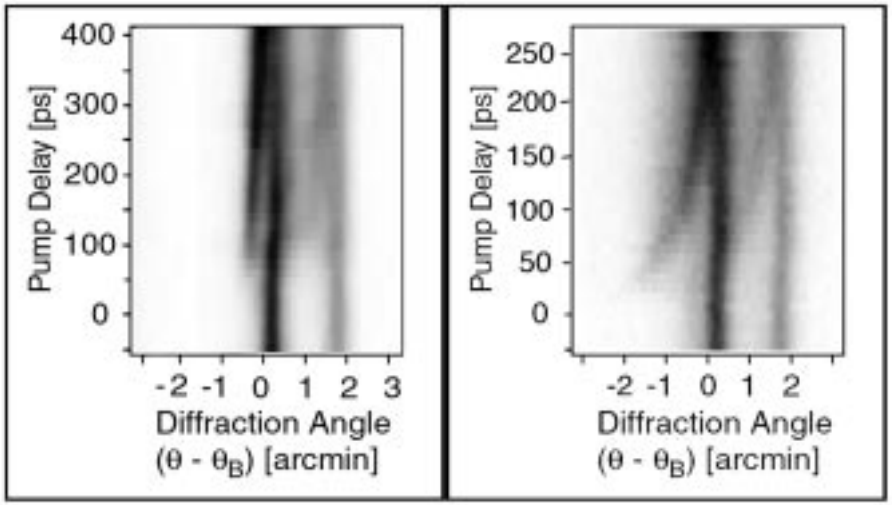


Ch. Rose-Petruck et al. *Nature* 398, 310 (1999).

Heating depths

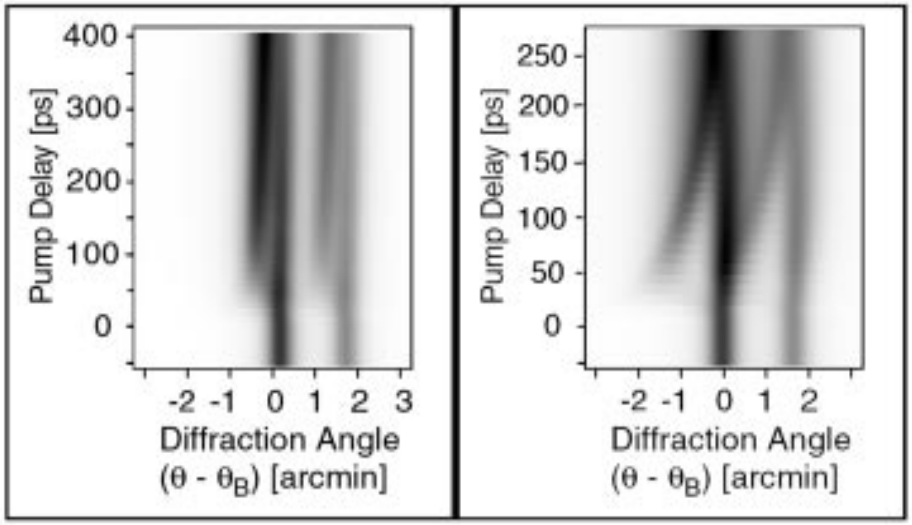
Experiment

Ge ***GaAs***



Theory

Ge ***GaAs***



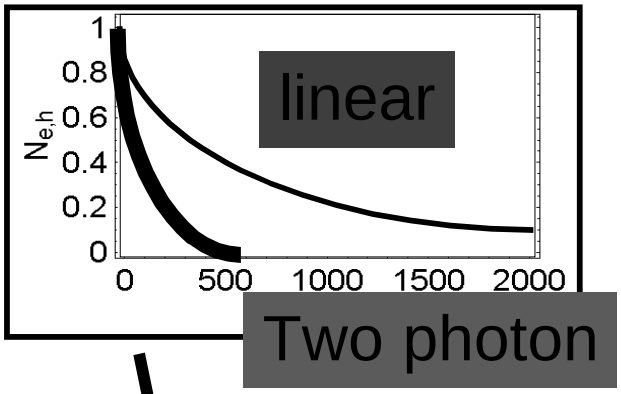
$\zeta = 1\ \mu m$ $\zeta = 275\ \mu m$

Heating depths

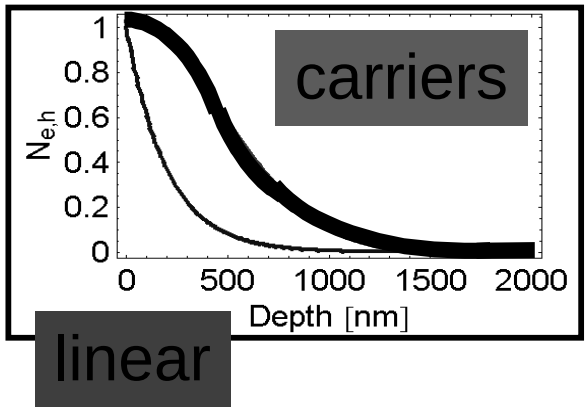
Two-temperature model

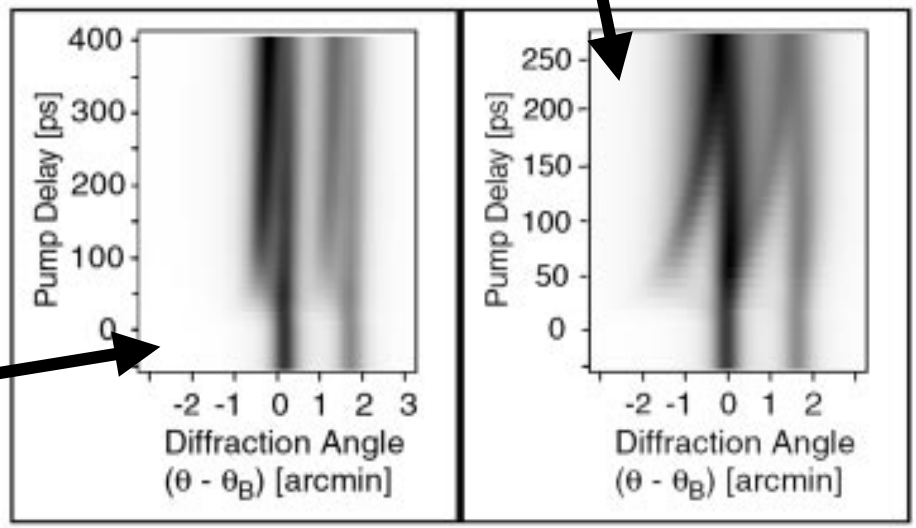
- ✓ *Two photon absorption*
- ✓ *High density carrier diffusion*
- ✓ *Auger recombination coefficients*

GaAs



Ge

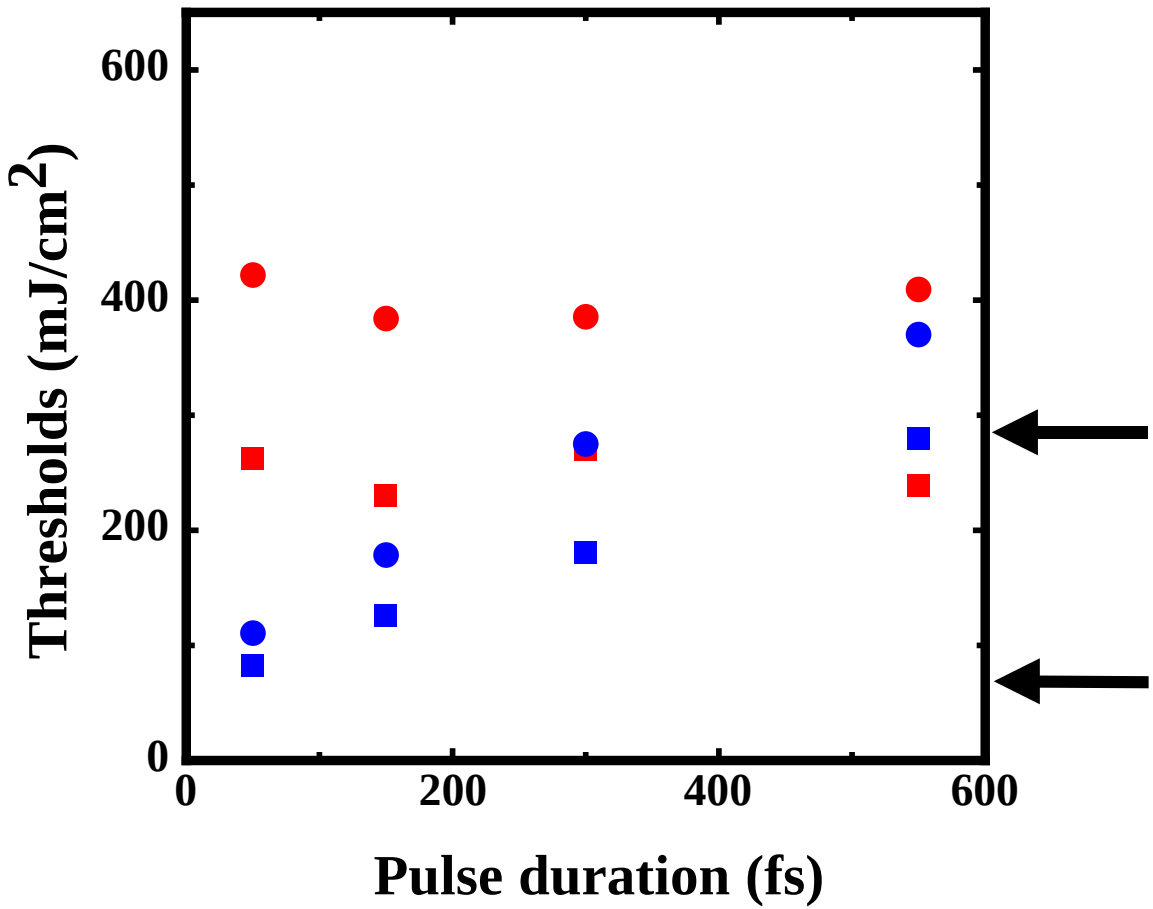




Dependence on pulse duration

Thermal Melting ■ ***GaAs*** ■ ***Ge***

Ablation ● ***GaAs*** ● ***Ge***



Calculated GaAs

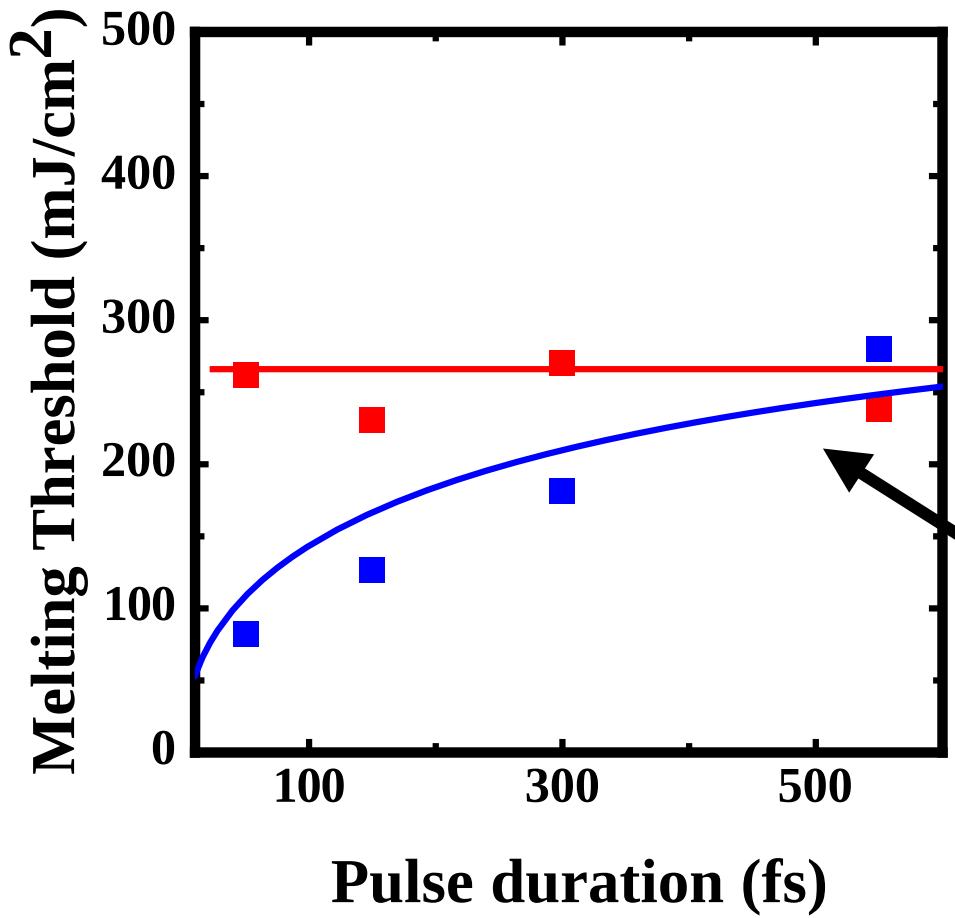
$\zeta = 1 / \alpha \approx 1\ \mu m$

Calculated Ge

$\zeta = 1 / \alpha \approx 200\ nm$

Dependence on pulse duration

Thermal Melting ■ ***GaAs*** ■ ***Ge***



Calculated Ge

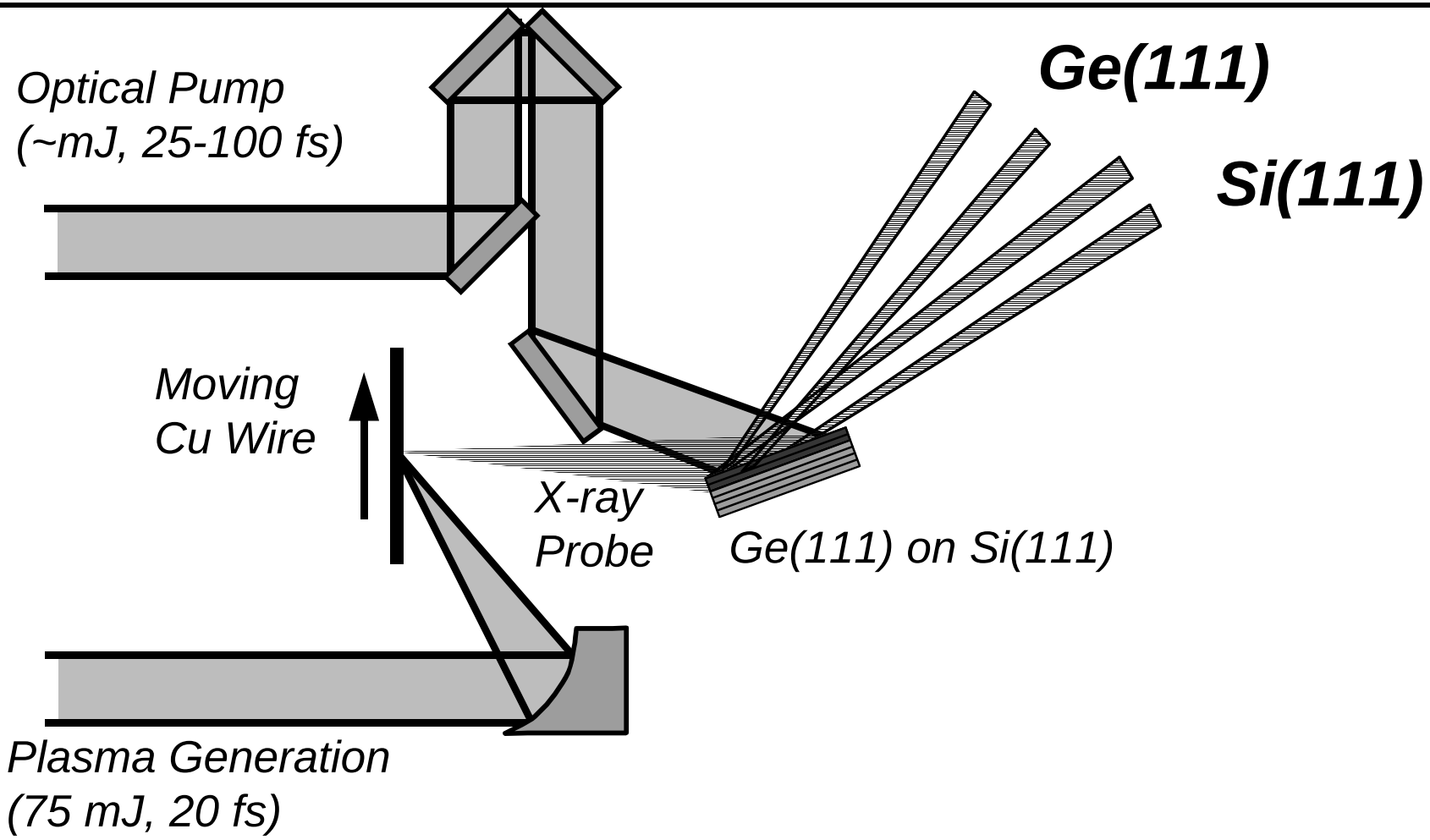
$\zeta = 1 / \alpha + (D\ \tau)^{1/2} \approx 1\ \mu m$

Calculated GaAs

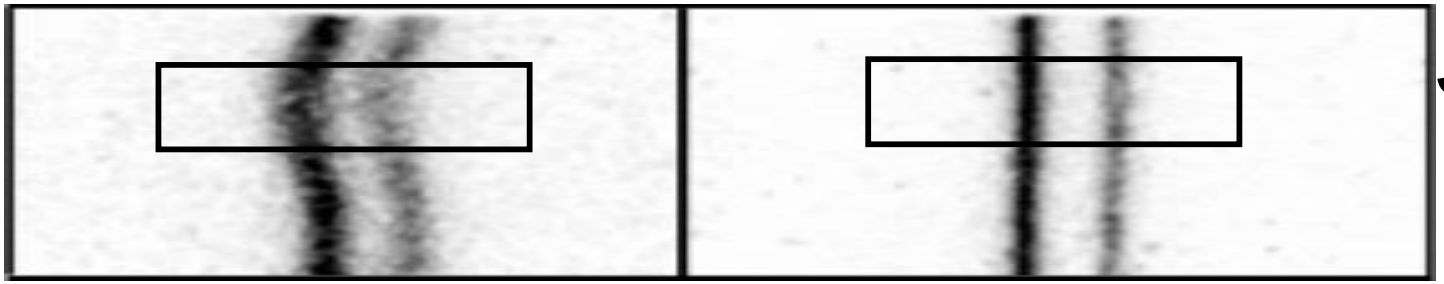
$\zeta = 1 / \alpha + \beta I$

A. Cavalleri et al., submitted

Probing Hetero-structures

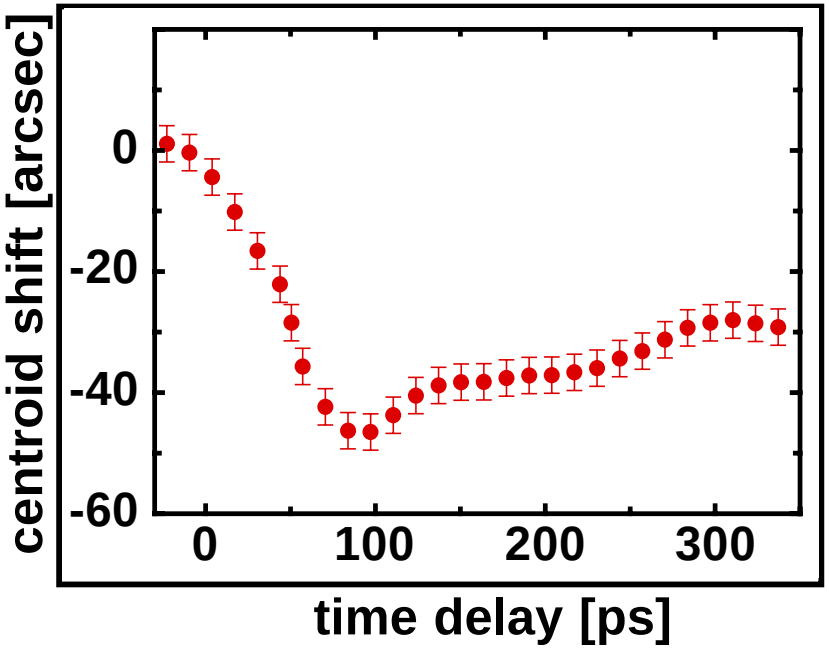


Centroid positions (400 nm film)

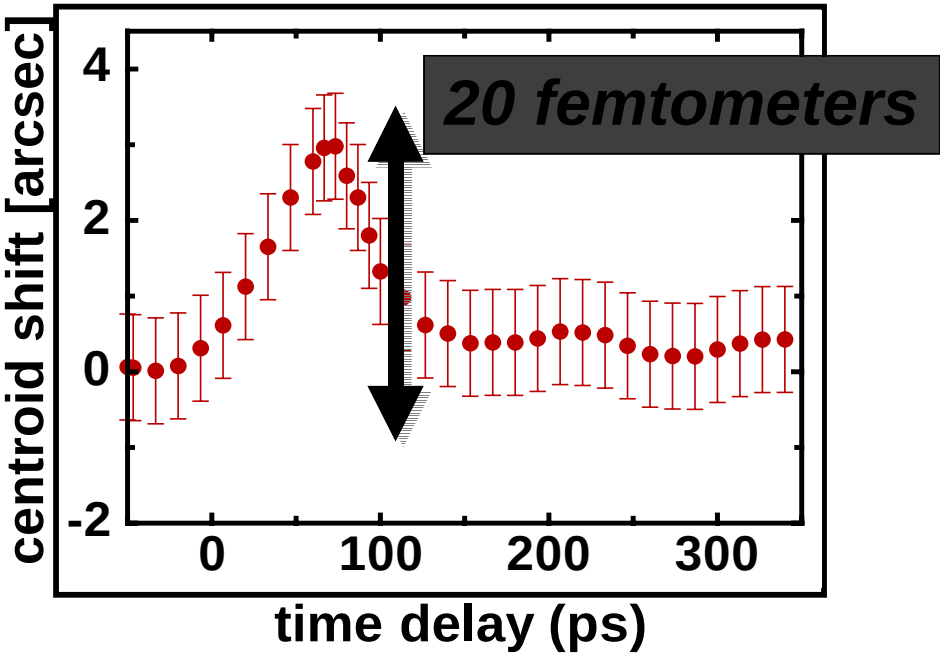
Ge  ***Si***

τ_{delay} **= 100 ps**

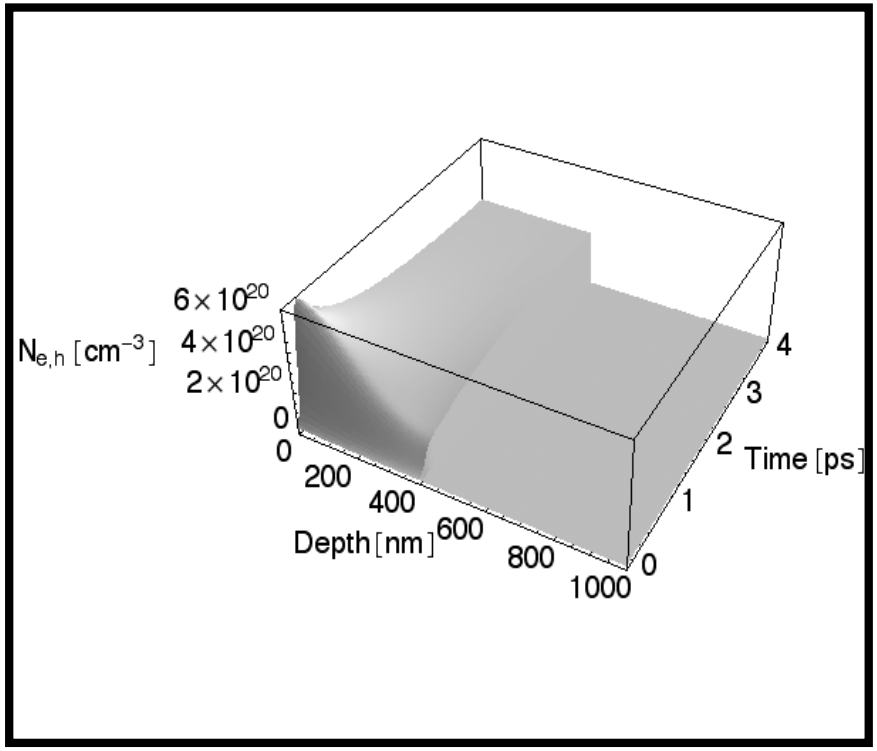
Expansion



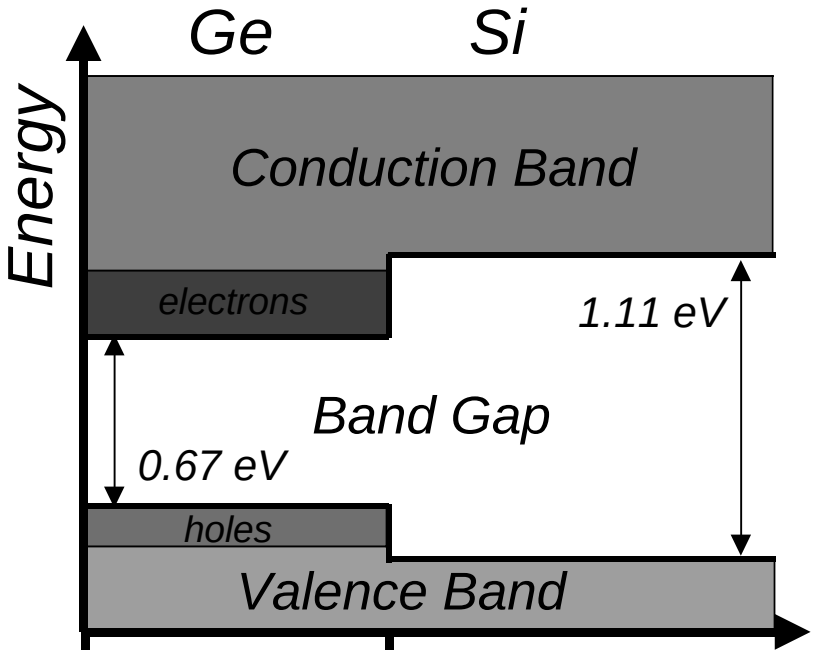
Compression



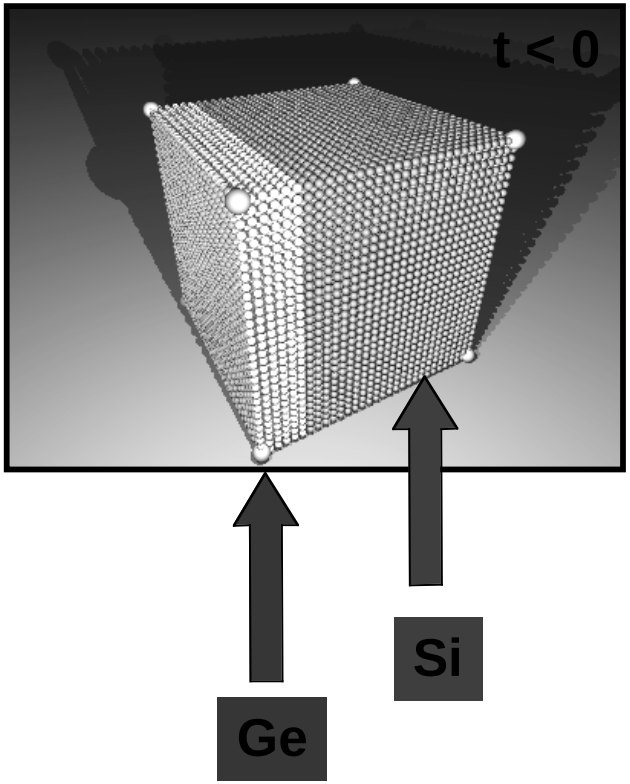
Carrier dynamics (400 nm)

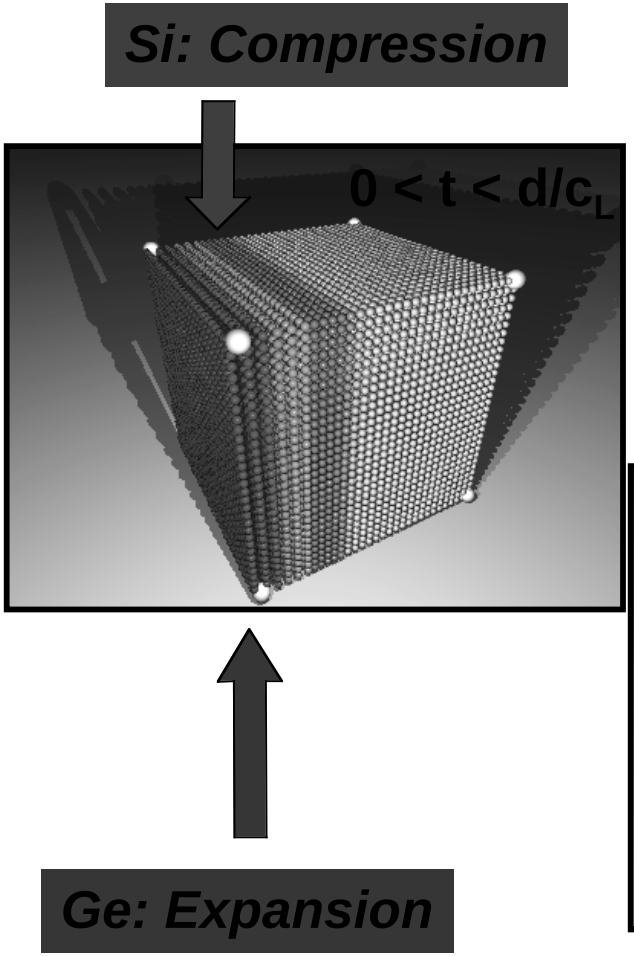


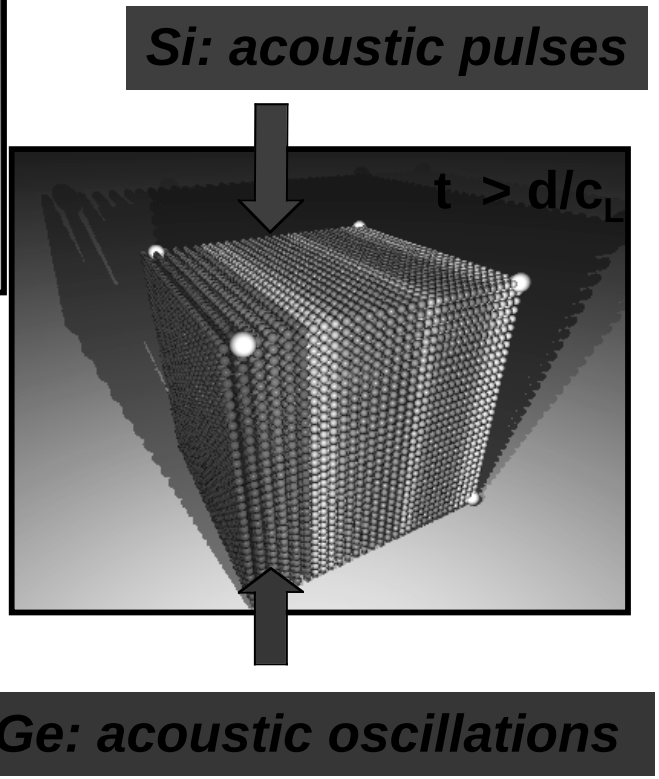
- ✓ ***Only Ge absorbs***
- ✓ ***Carrier diffusion***
- ✓ ***Carriers are confined***



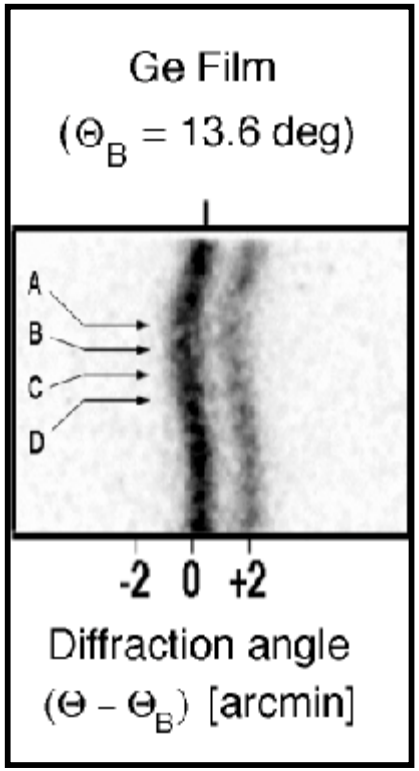
Coherent Acoustic Phonons: Films

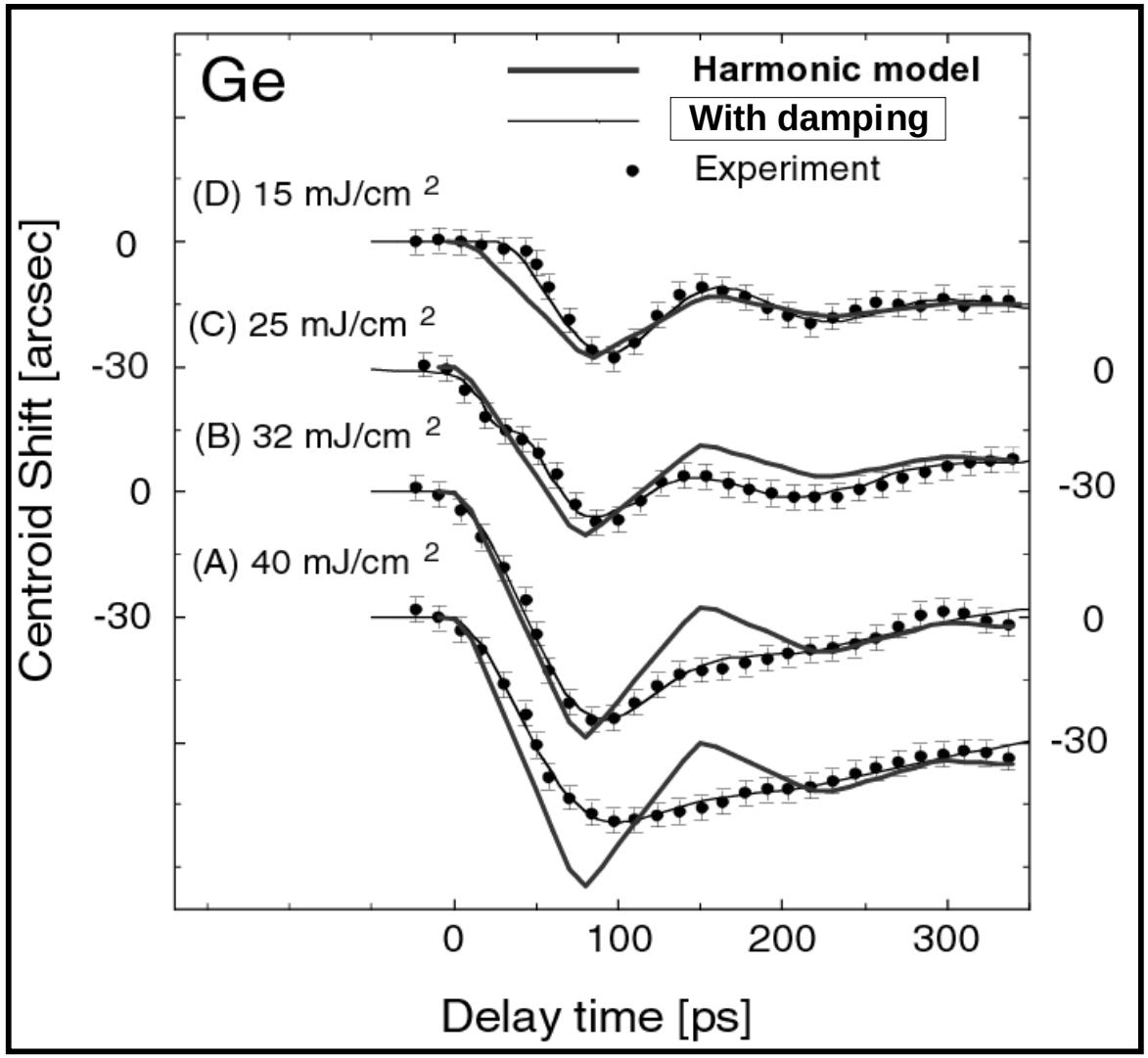




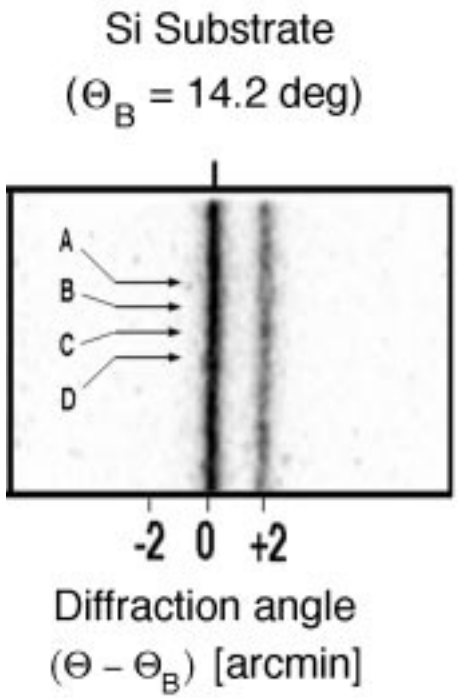


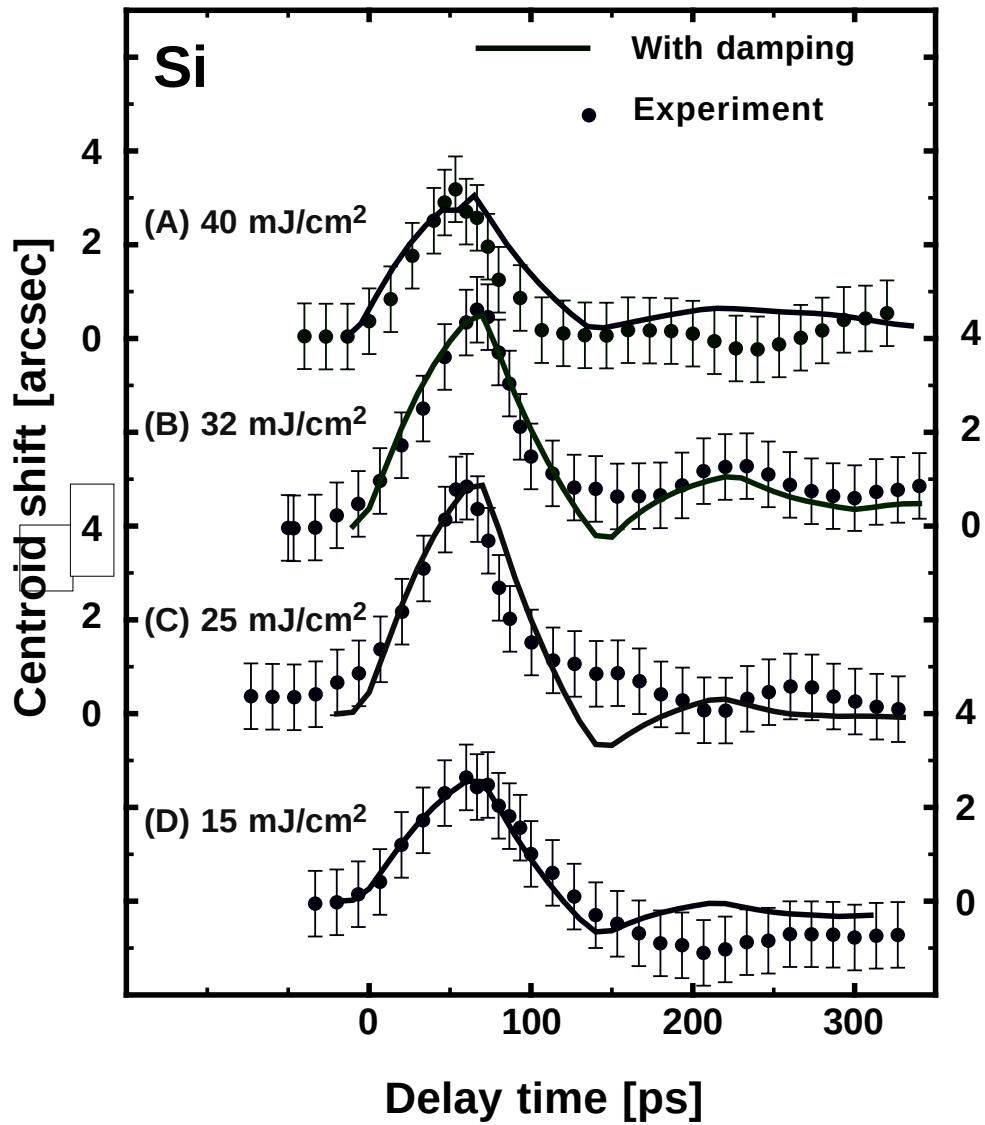
Fluence Dependent Damping



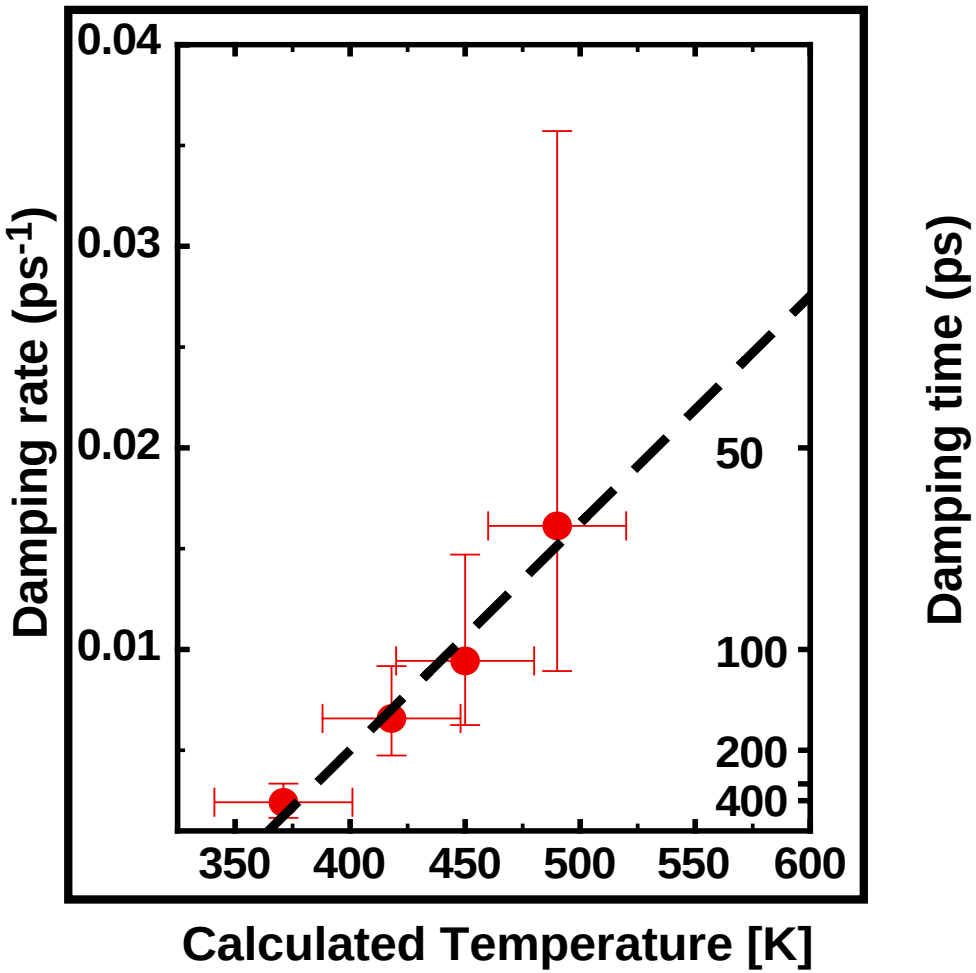


Fluence Dependent Damping

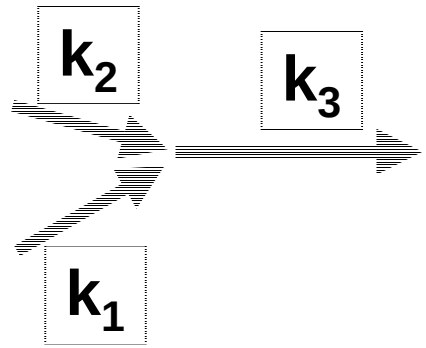




Coherent Phonon Damping Times

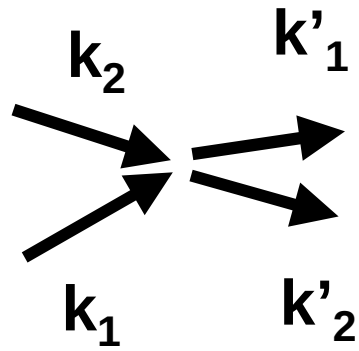


Phonon collision



T_1~ 100 ns

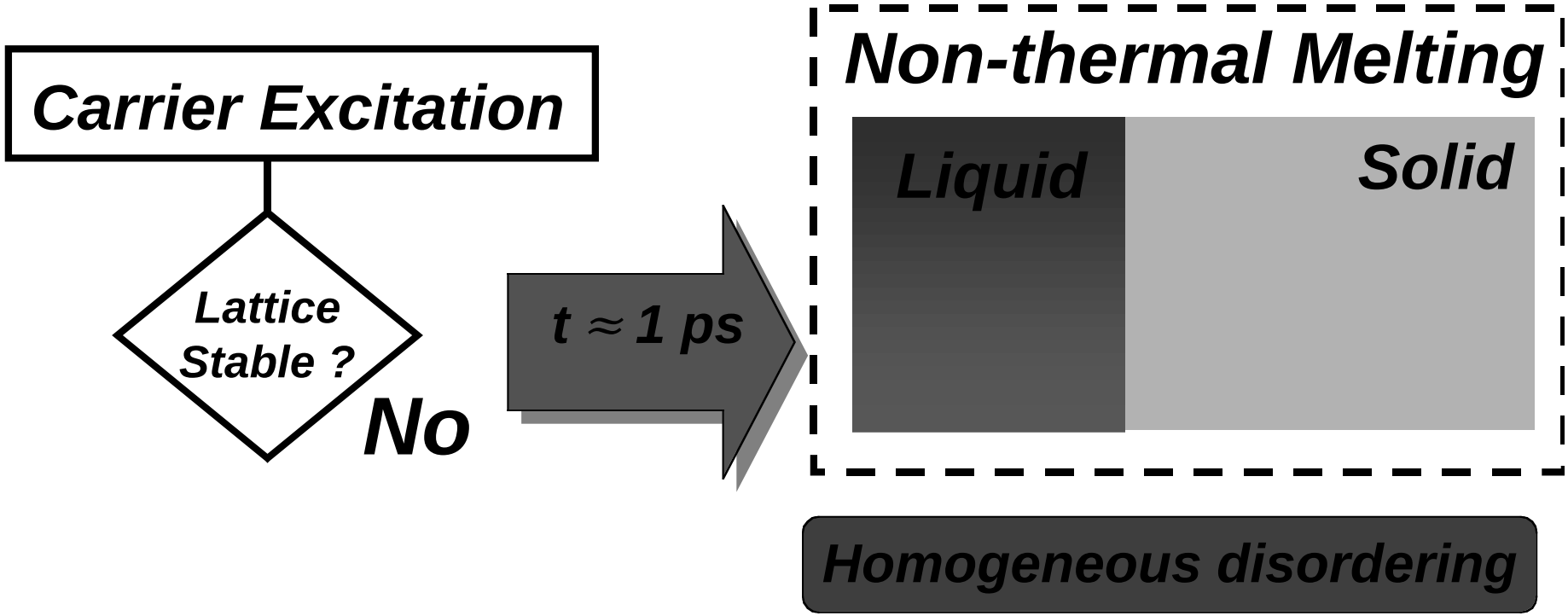
Phonon dephasing



T_2 ~ 100 ps

A. Cavalleri et al., *Phys. Rev. Lett.* 85, 586 (2000)

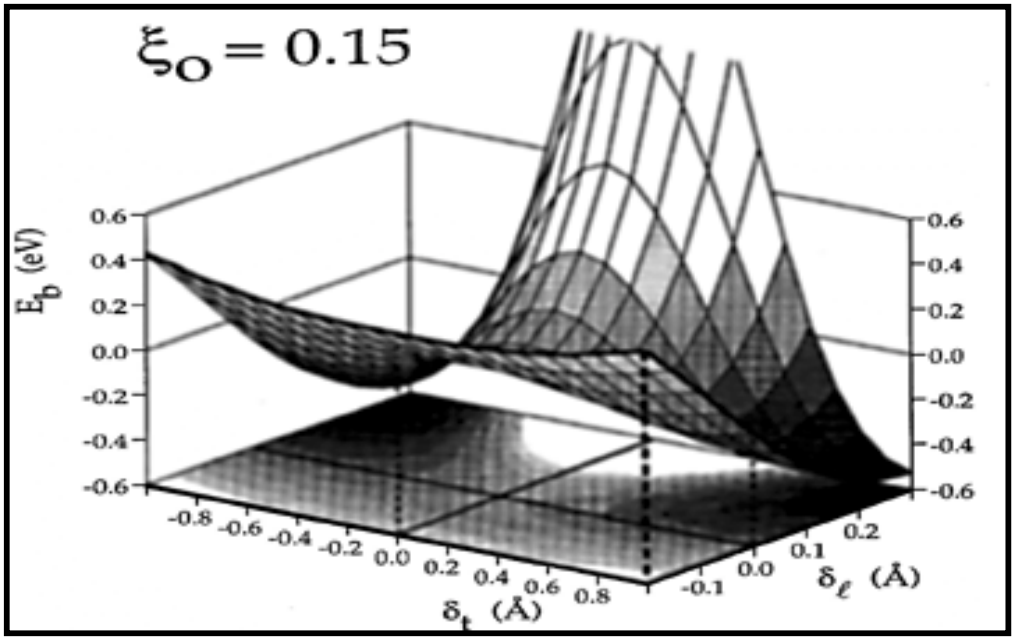
Ultrafast Melting Dynamics



J.A. van Vechten et al., *Phys.* Lett. *A* *74A*, *422 (1979).*

P.L. Silvestrelli et al., *Phys. Rev.* Lett. *77*, *3149 (1996).*

Potential Energy Surface

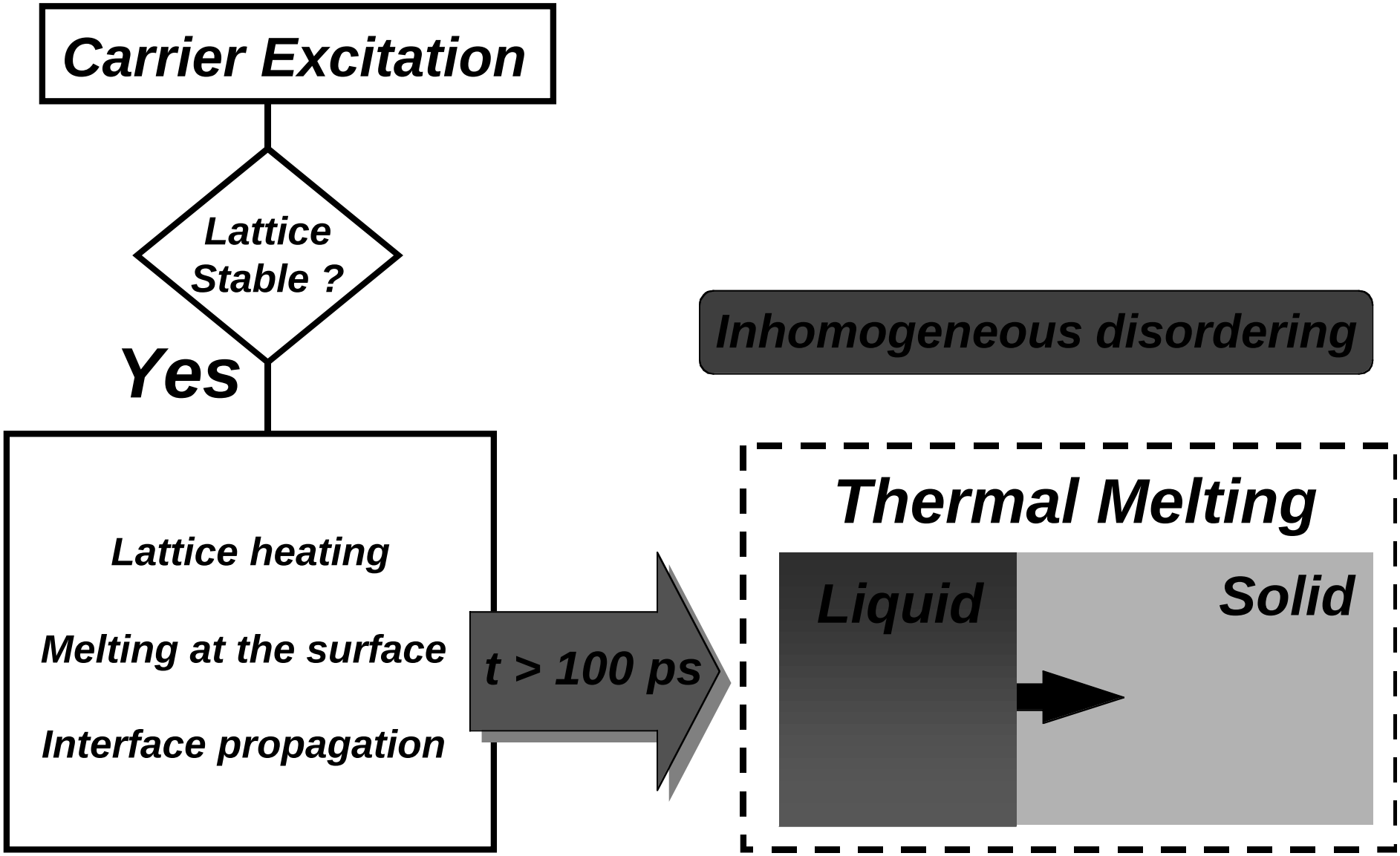


10^{22} excited carriers/cm^3
25% valence electrons
in the conduction band

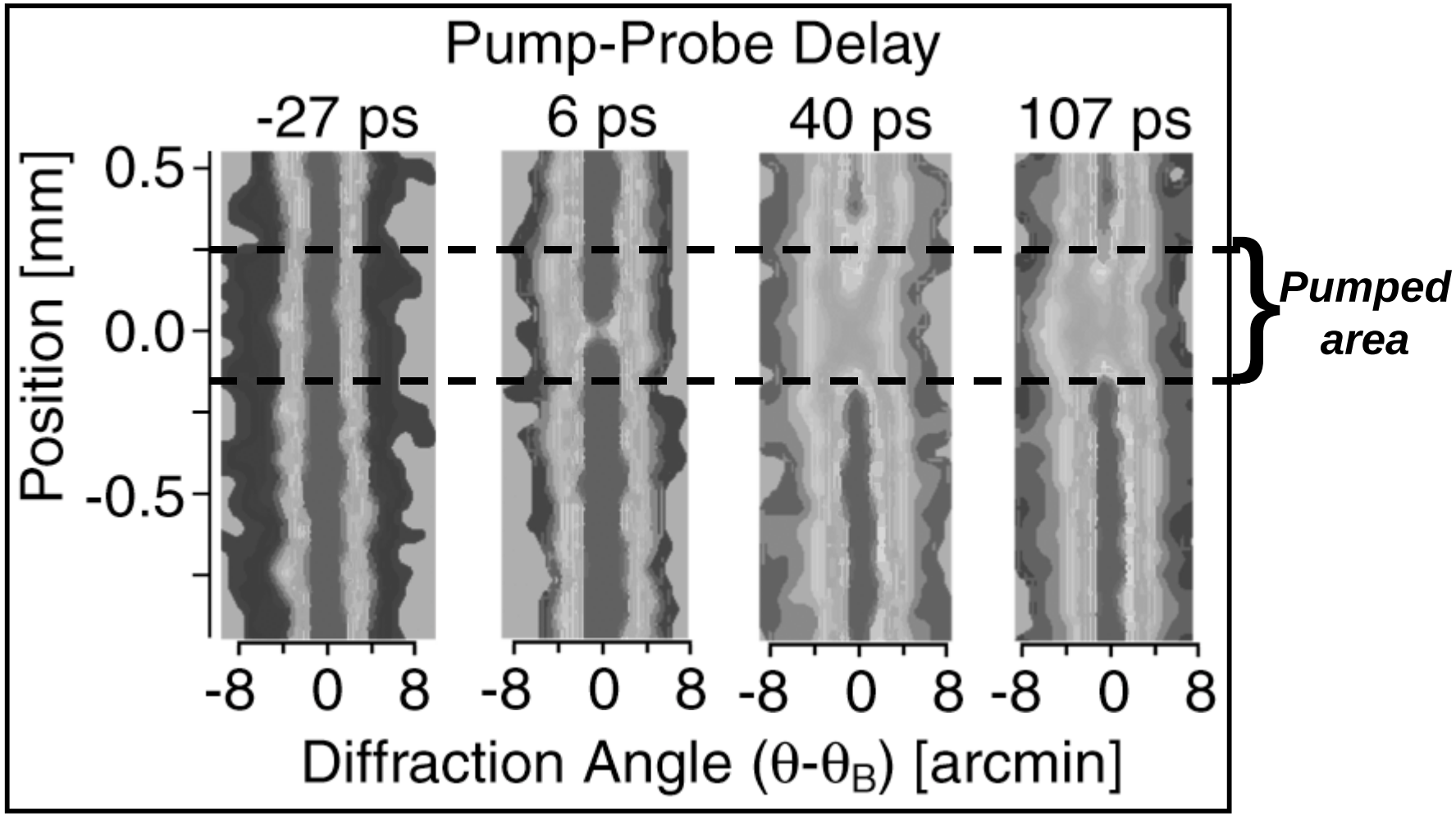
Unstable lattice

P. Stampfli et al., *Phys. Rev.* B *49*, *7299 (1994).*

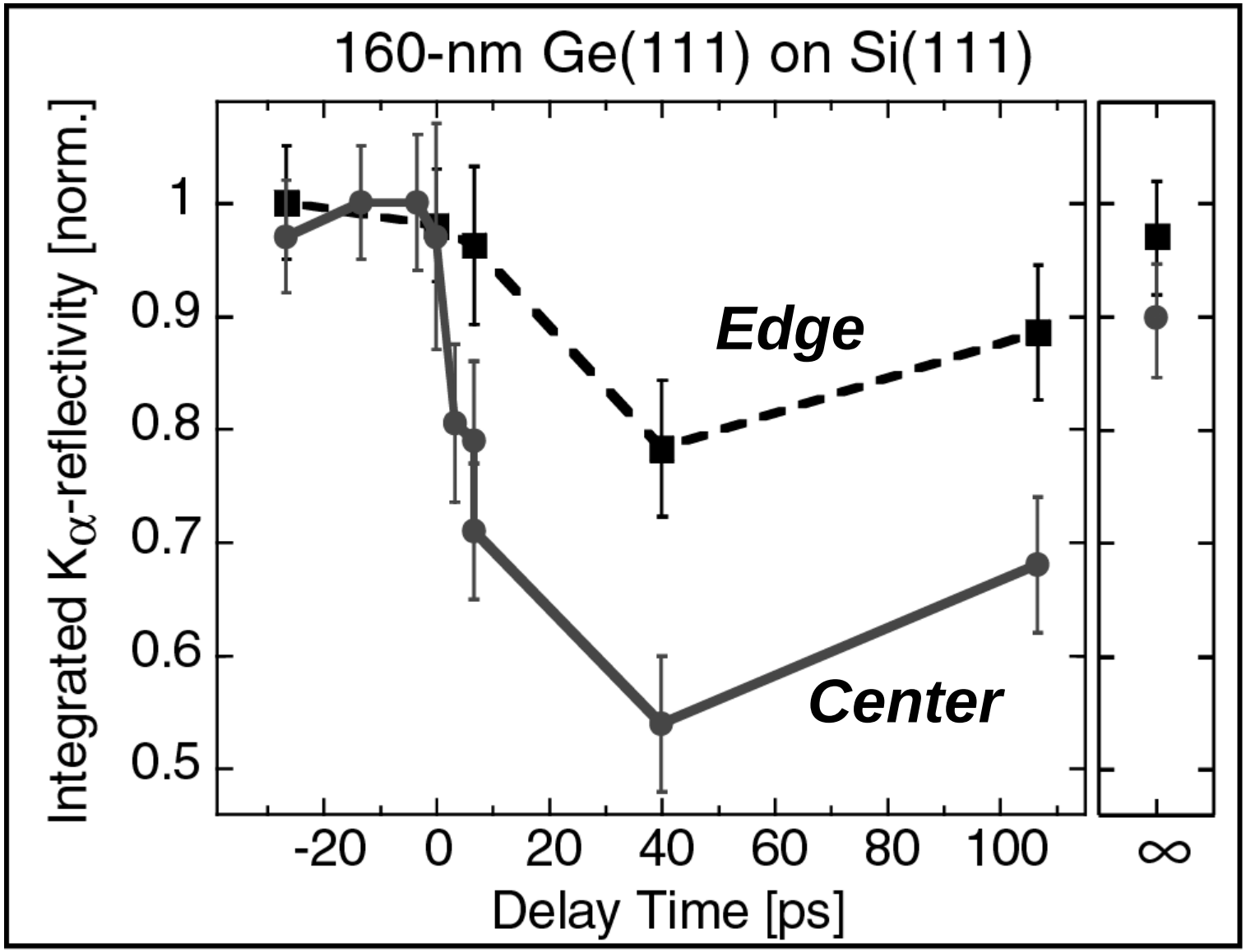
Ultrafast Melting Dynamics



Ultrafast Melting in 160 nm Ge



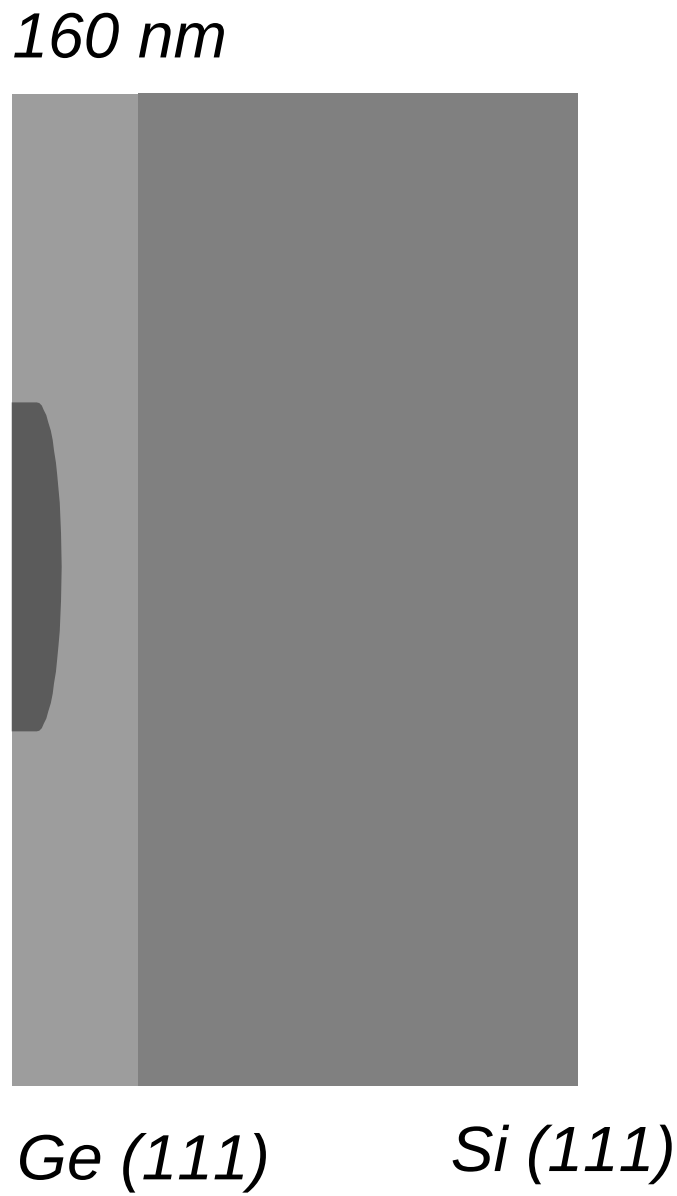
Ultrafast Melting: Integrated K_α Reflectivity



Ultrafast Melting

40 nm in < 7 ps

Homogeneous Disordering

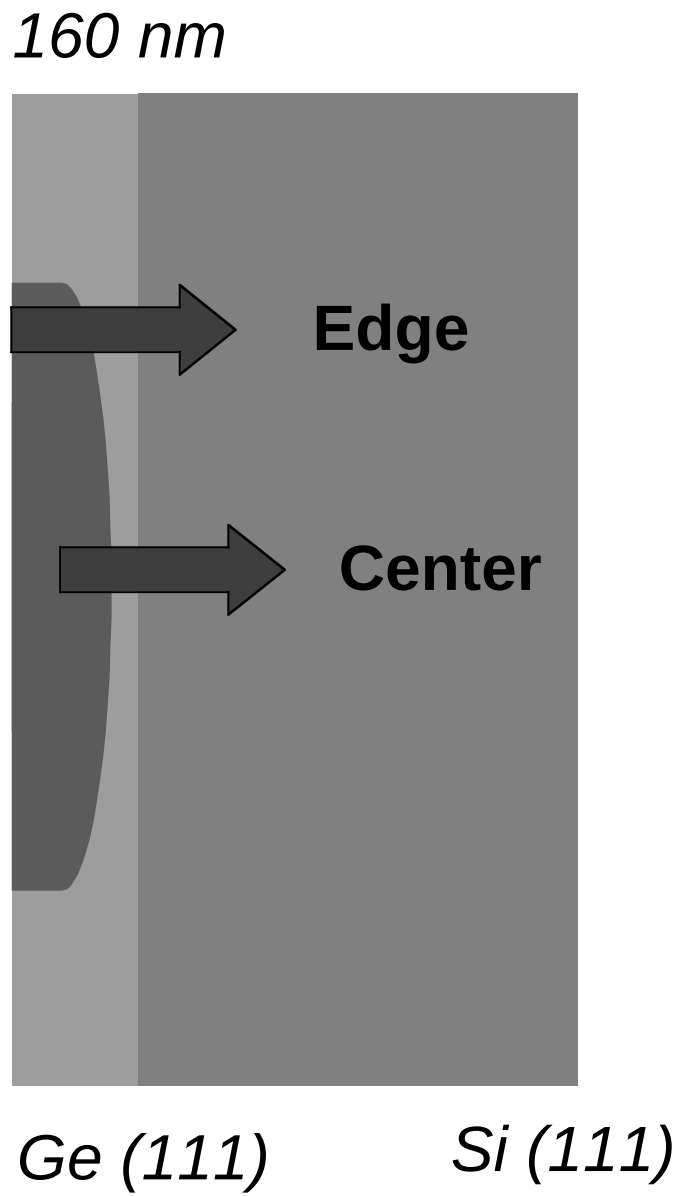


Ultrafast Melting

Inhomogeneous

Highly superheated solid

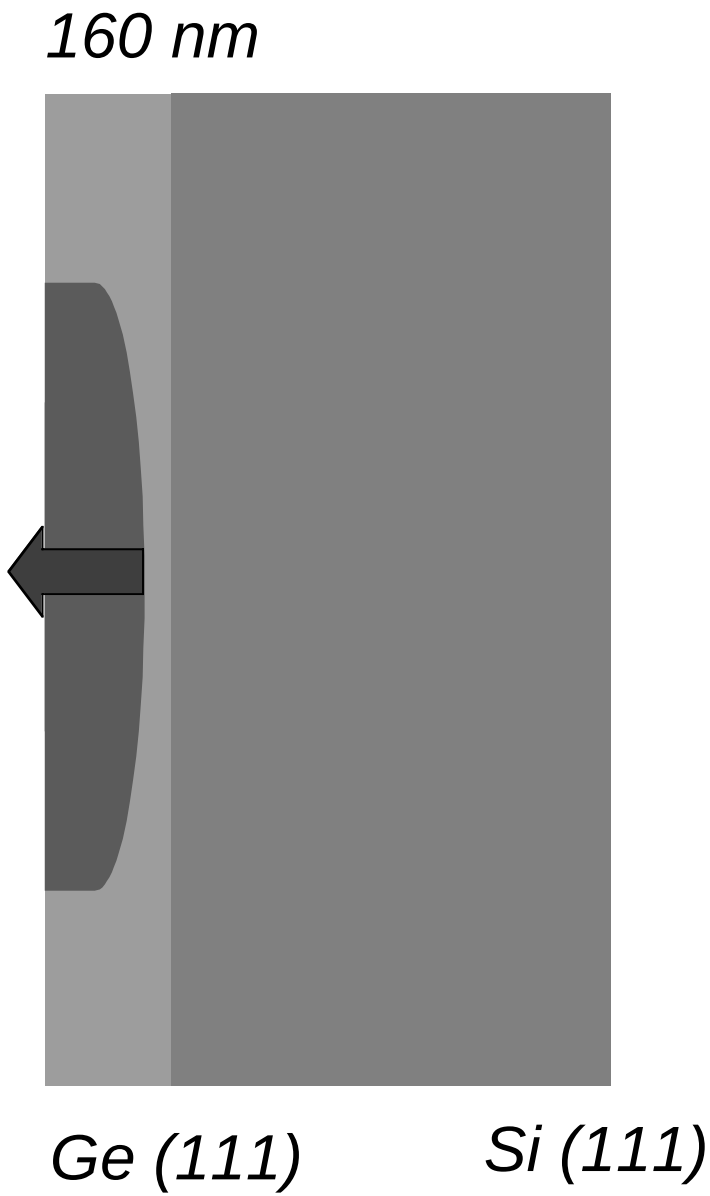
Interface velocity
V < 700 m / sec



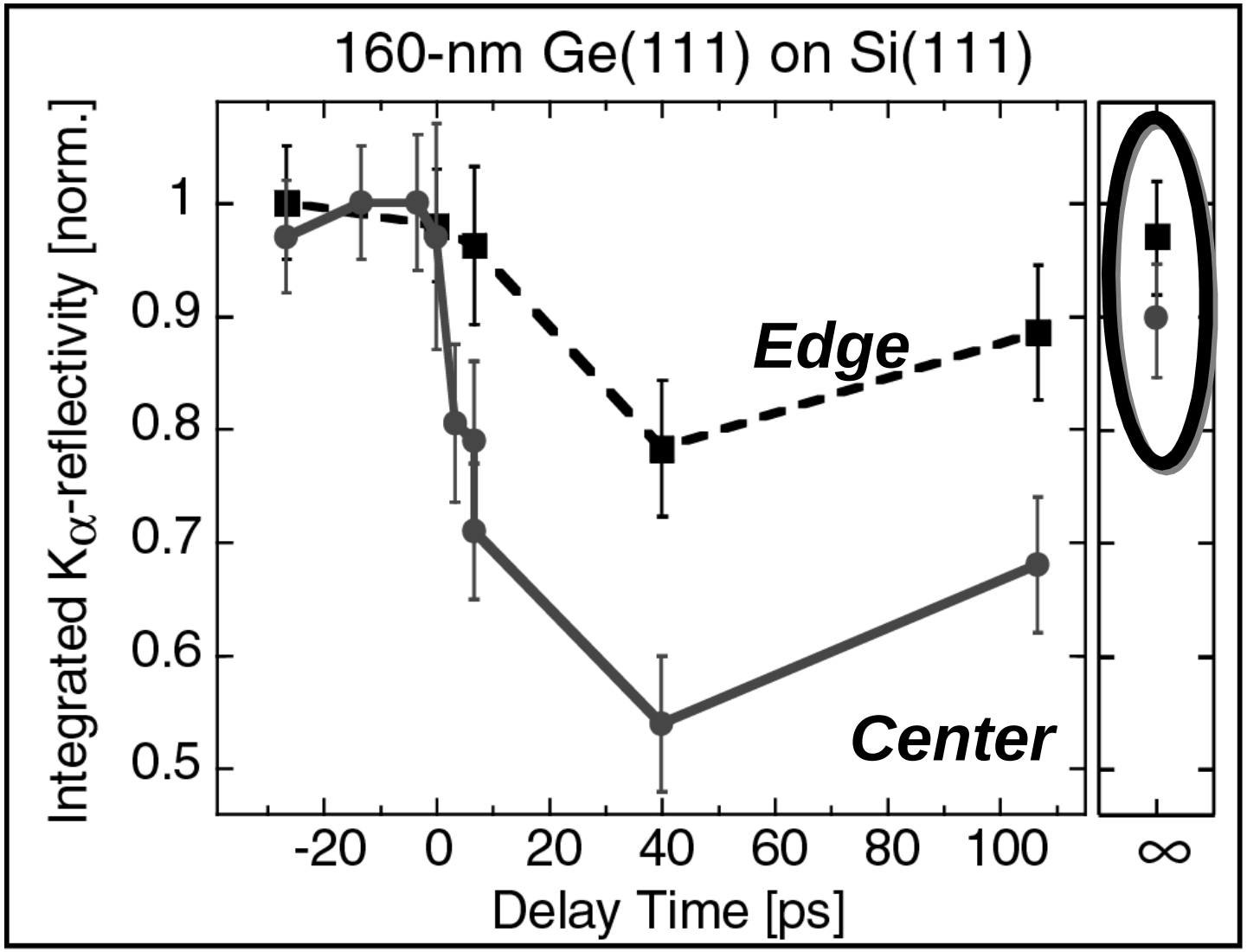
Ultrafast Melting

Re-solidification

Solid-liquid-solid



Ultrafast Melting: Integrated K_α Reflectivity

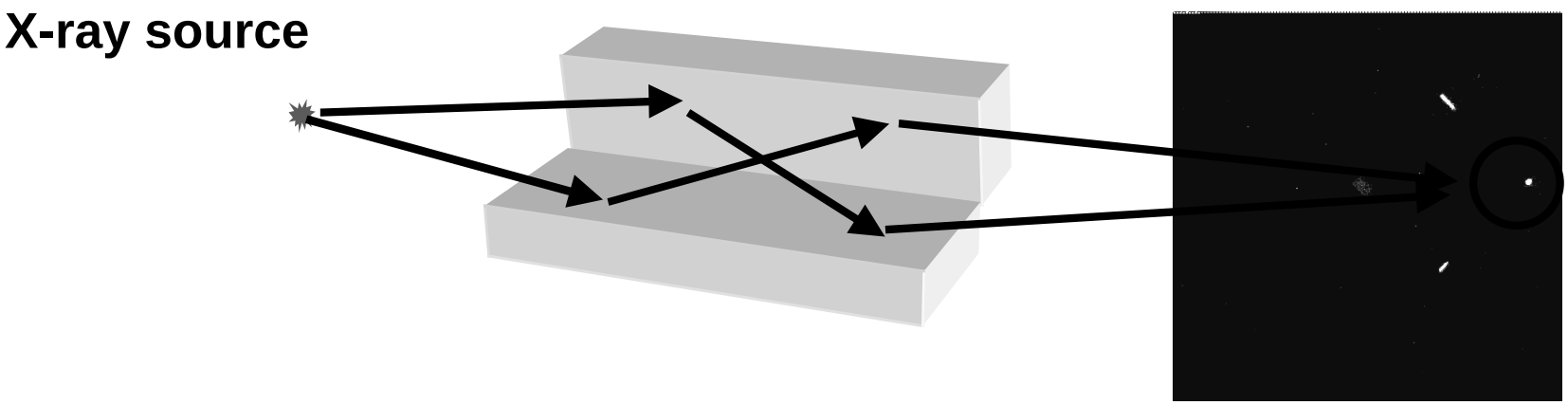


C.W. Siders, et al., *Science* 286, 1340, (1999).

X-ray Focusing

Elliptical surfaces

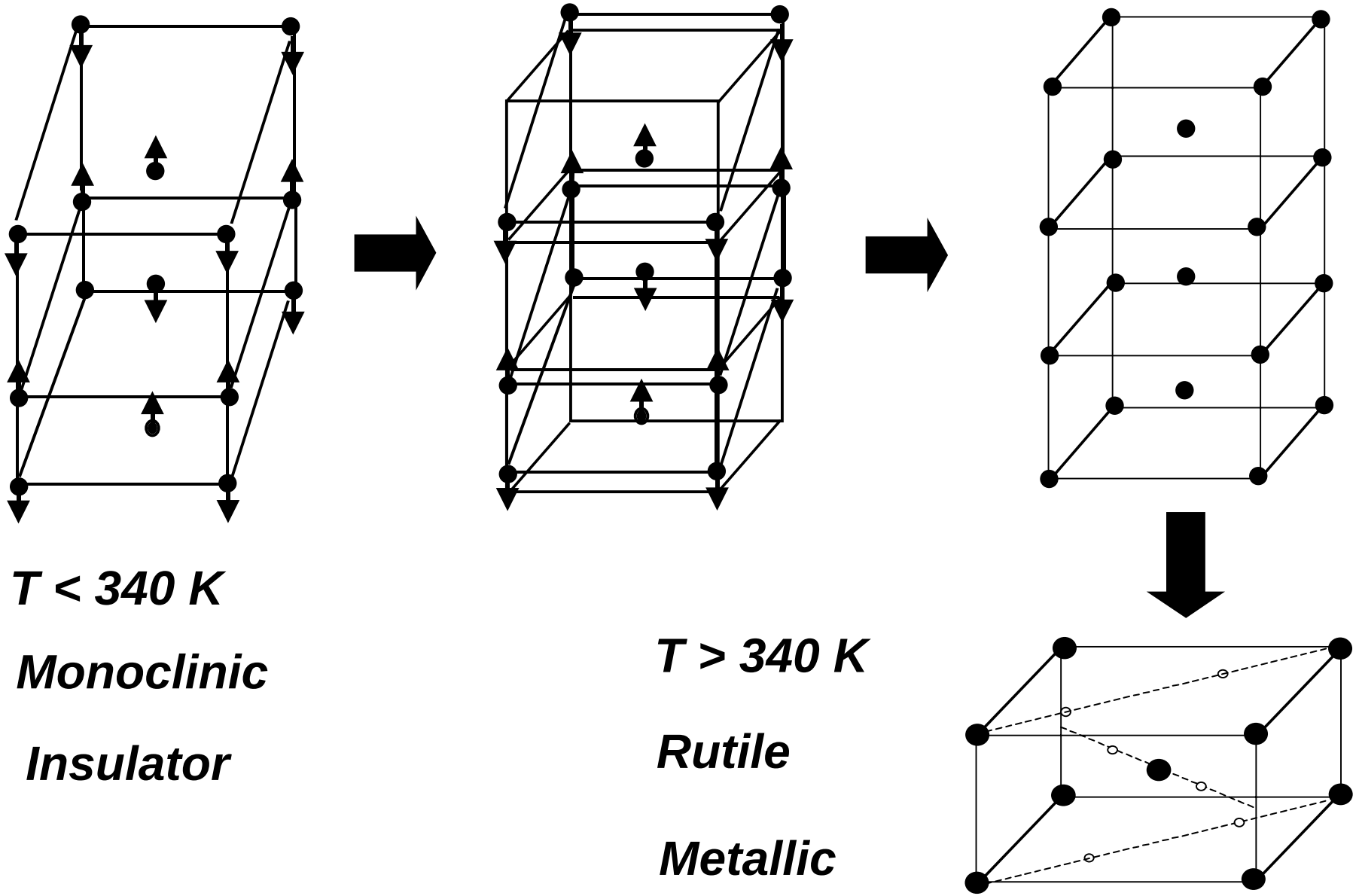
with graded Bragg coating



Spot diameter = 54 μm

2000 photons / shot

Structural Transition



T < 340 K

Monoclinic

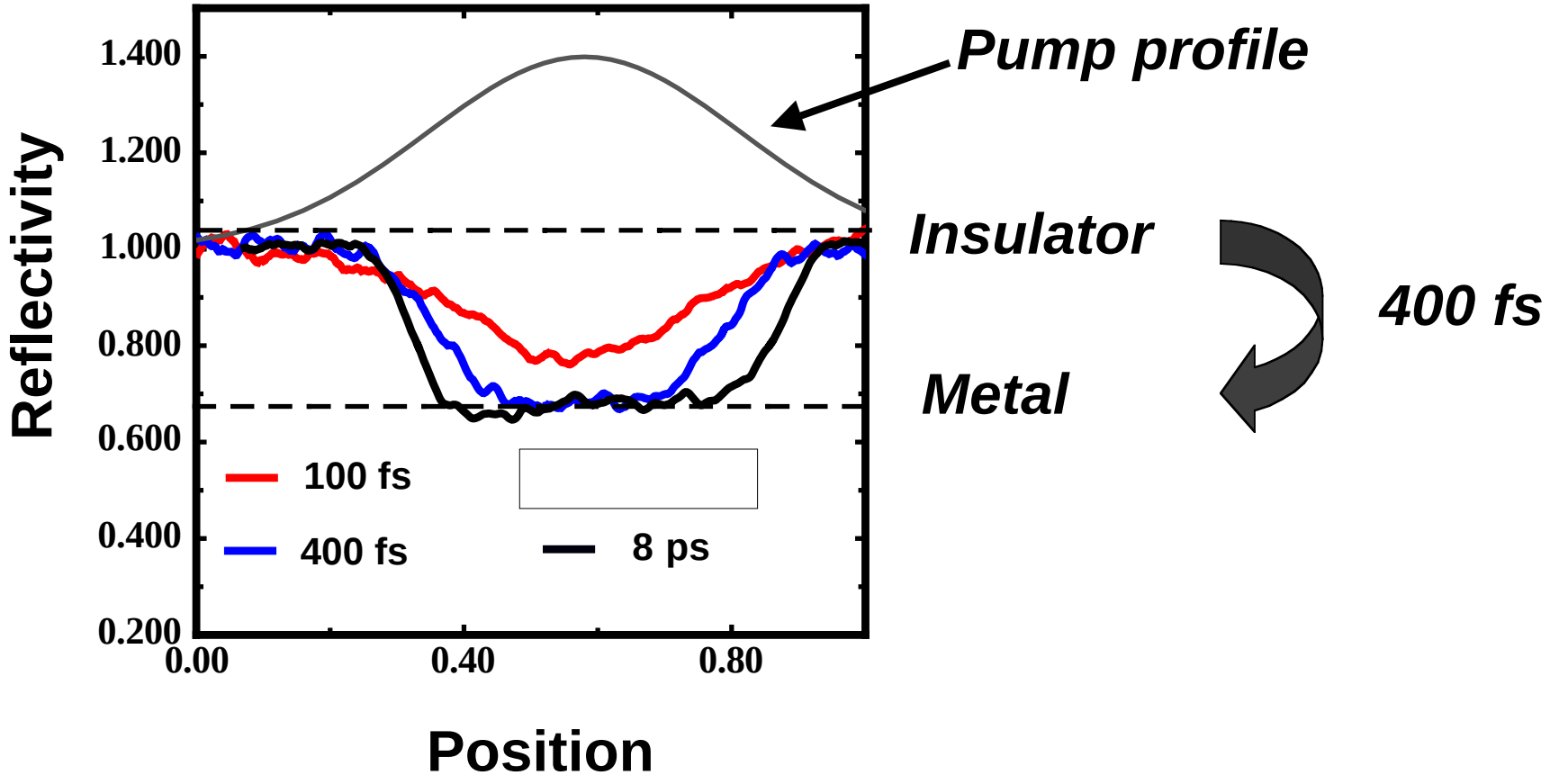
Insulator

T > 340 K

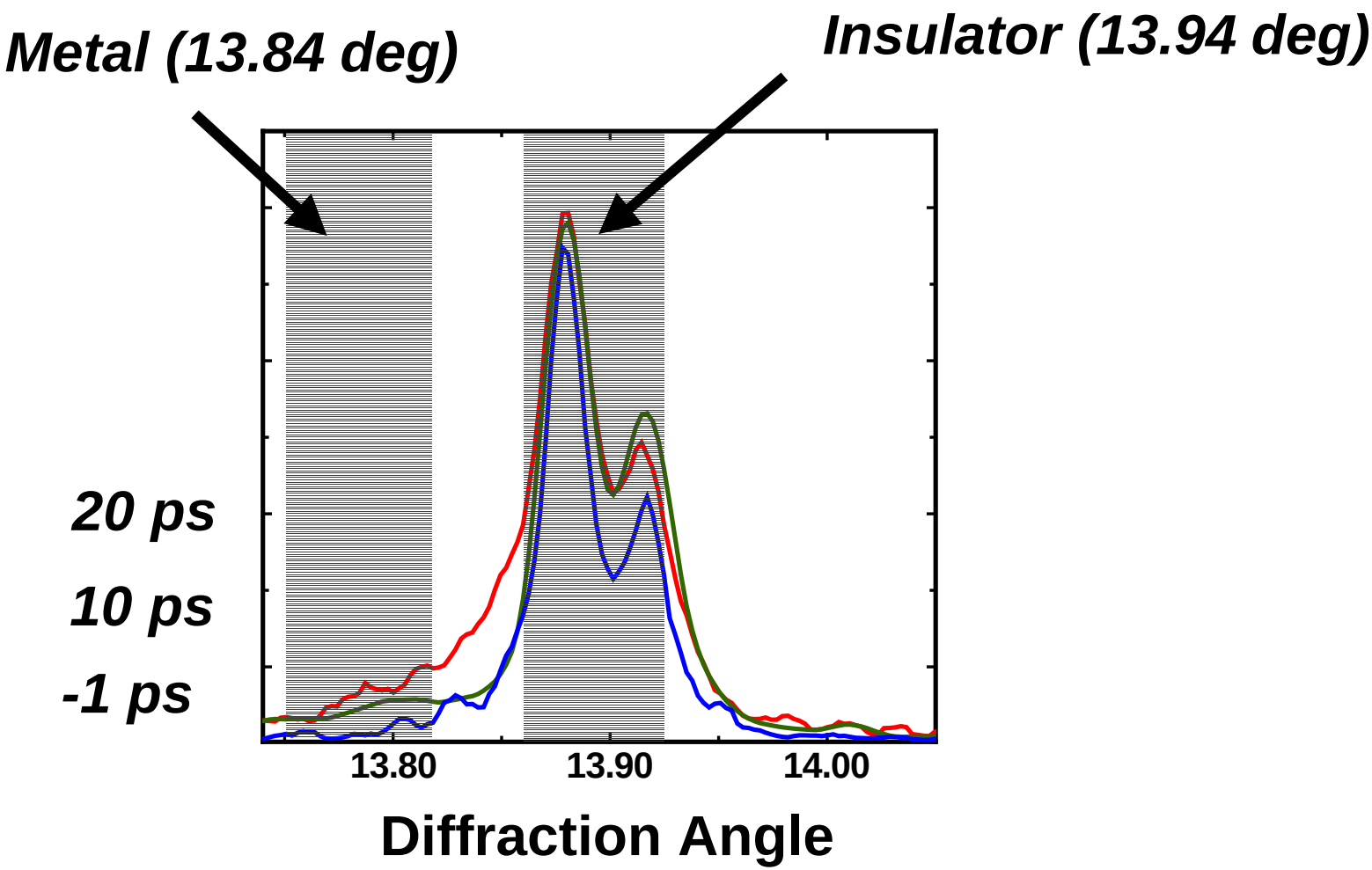
Rutile

Metallic

Ultrafast Optical Data: Insulator to Metal

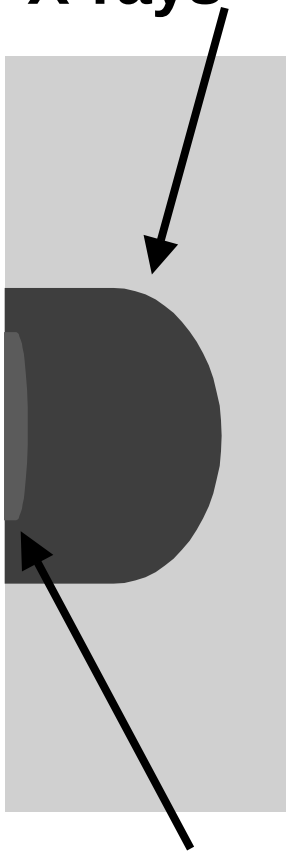


Ultrafast X-ray diffraction: bulk VO_2 (110)

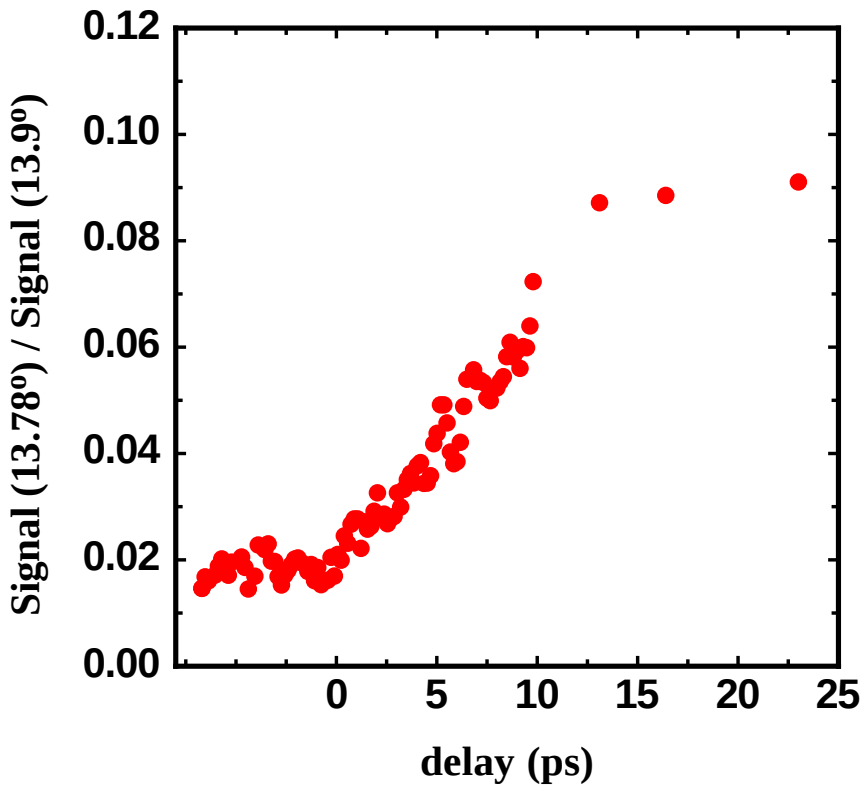


Time response

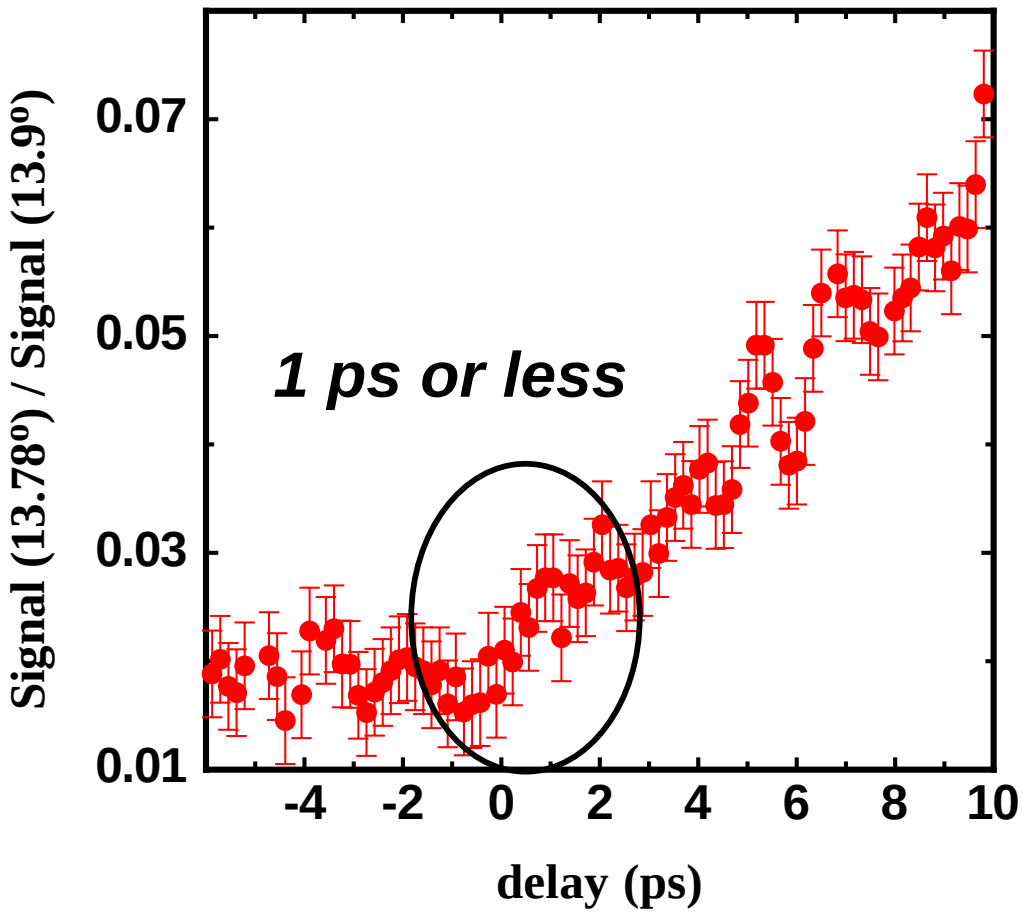
X-rays ≈ 3000 nm



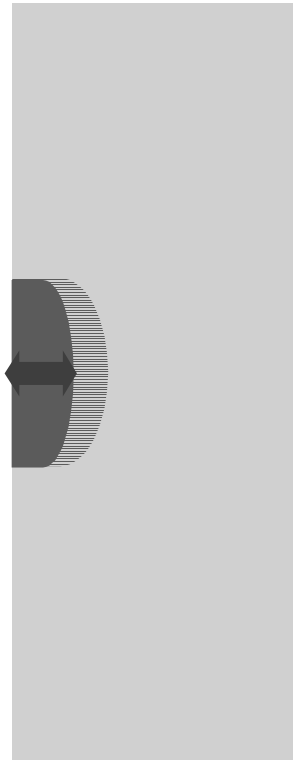
Metallic phase ≈ 300 nm



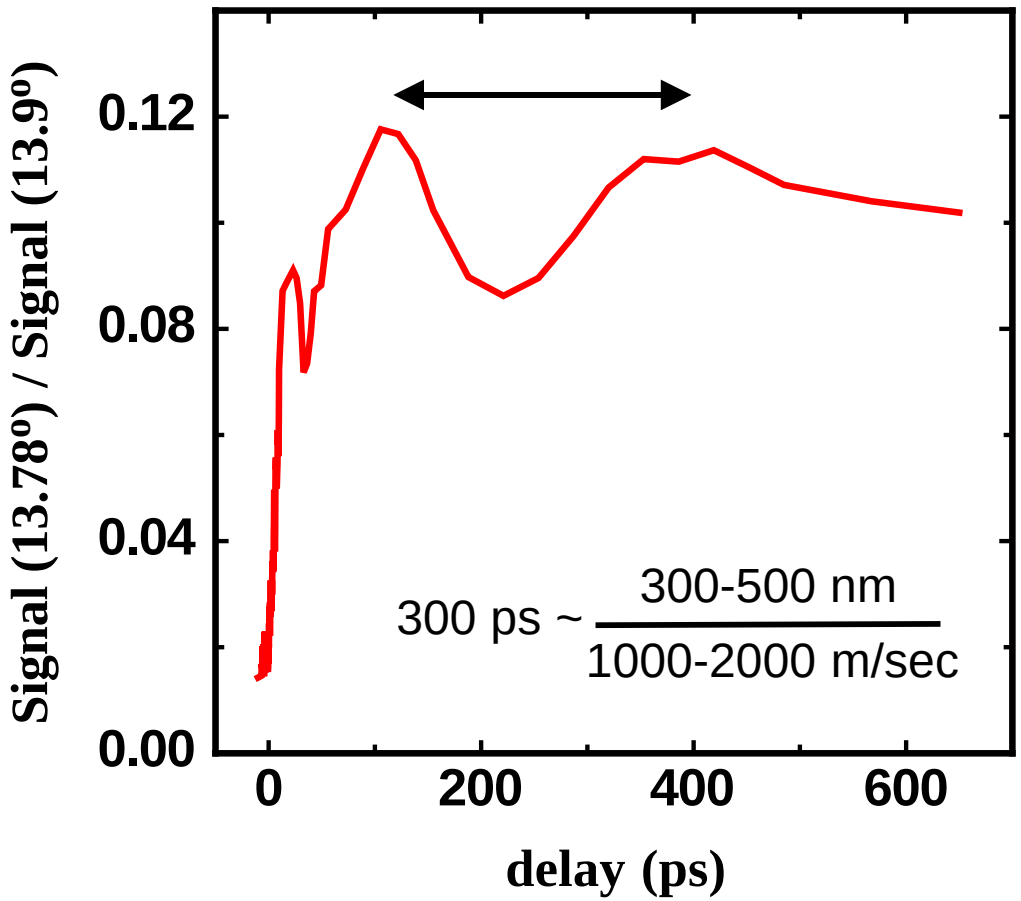
Time response



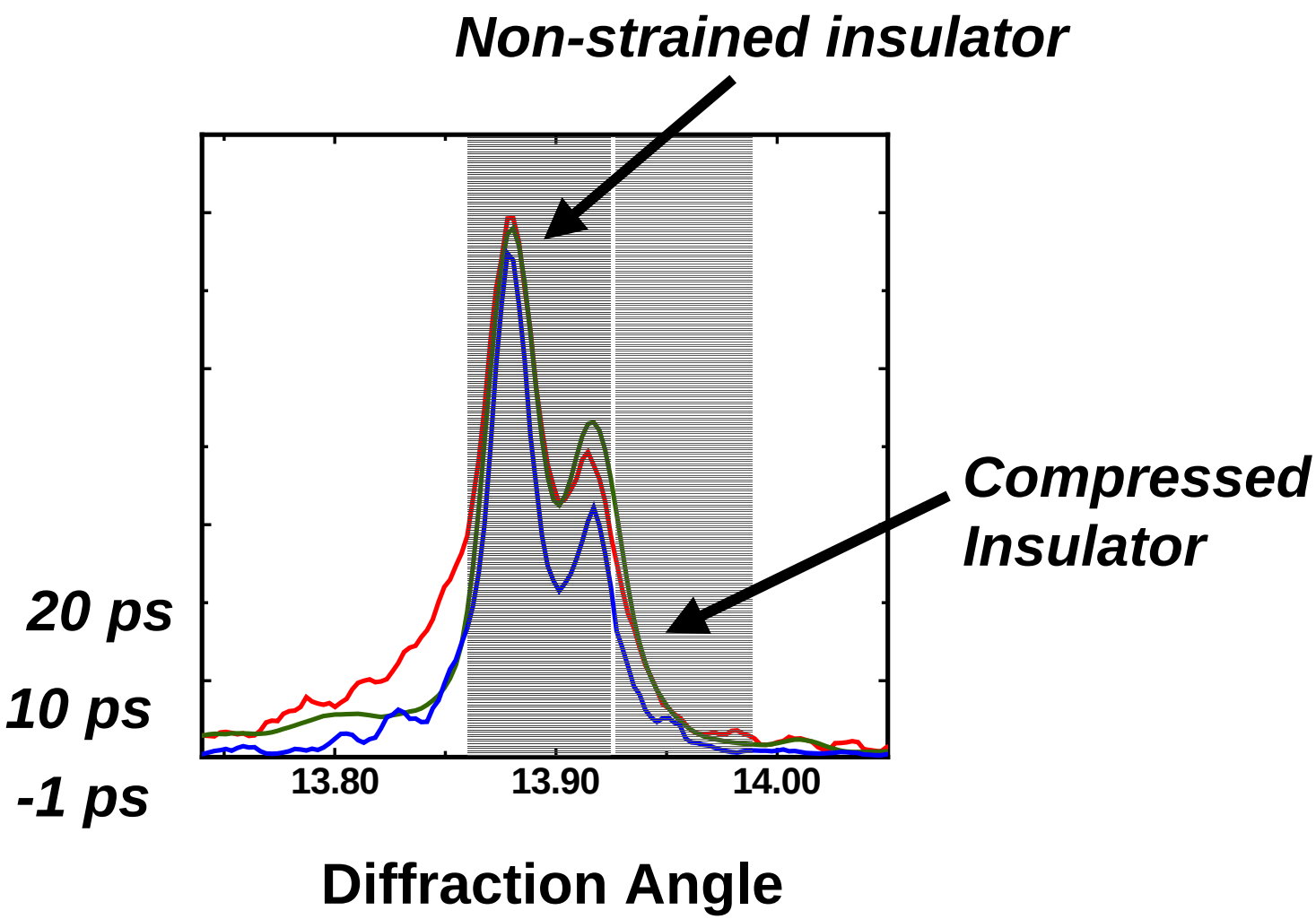
Long time response



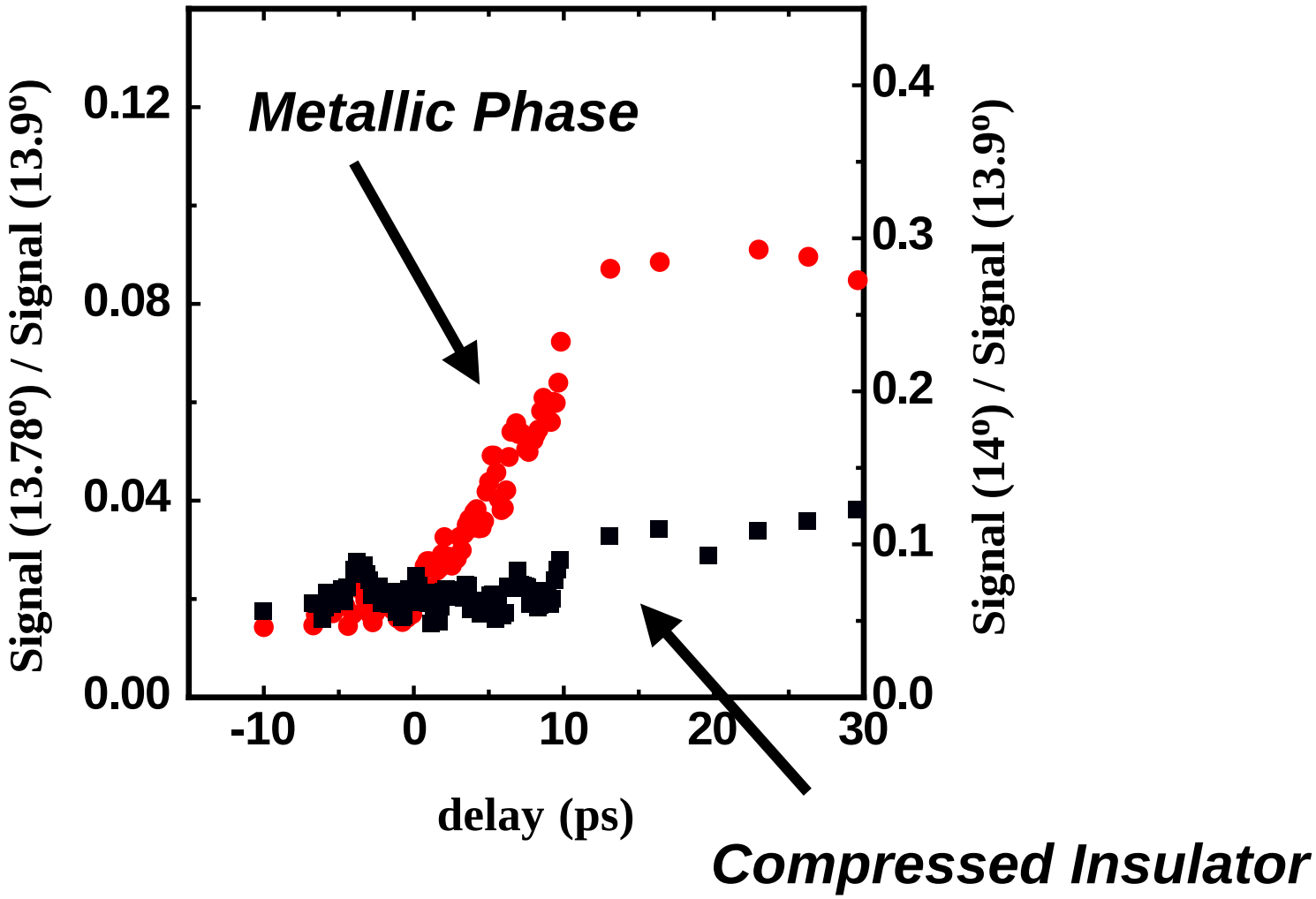
Acoustic response ?



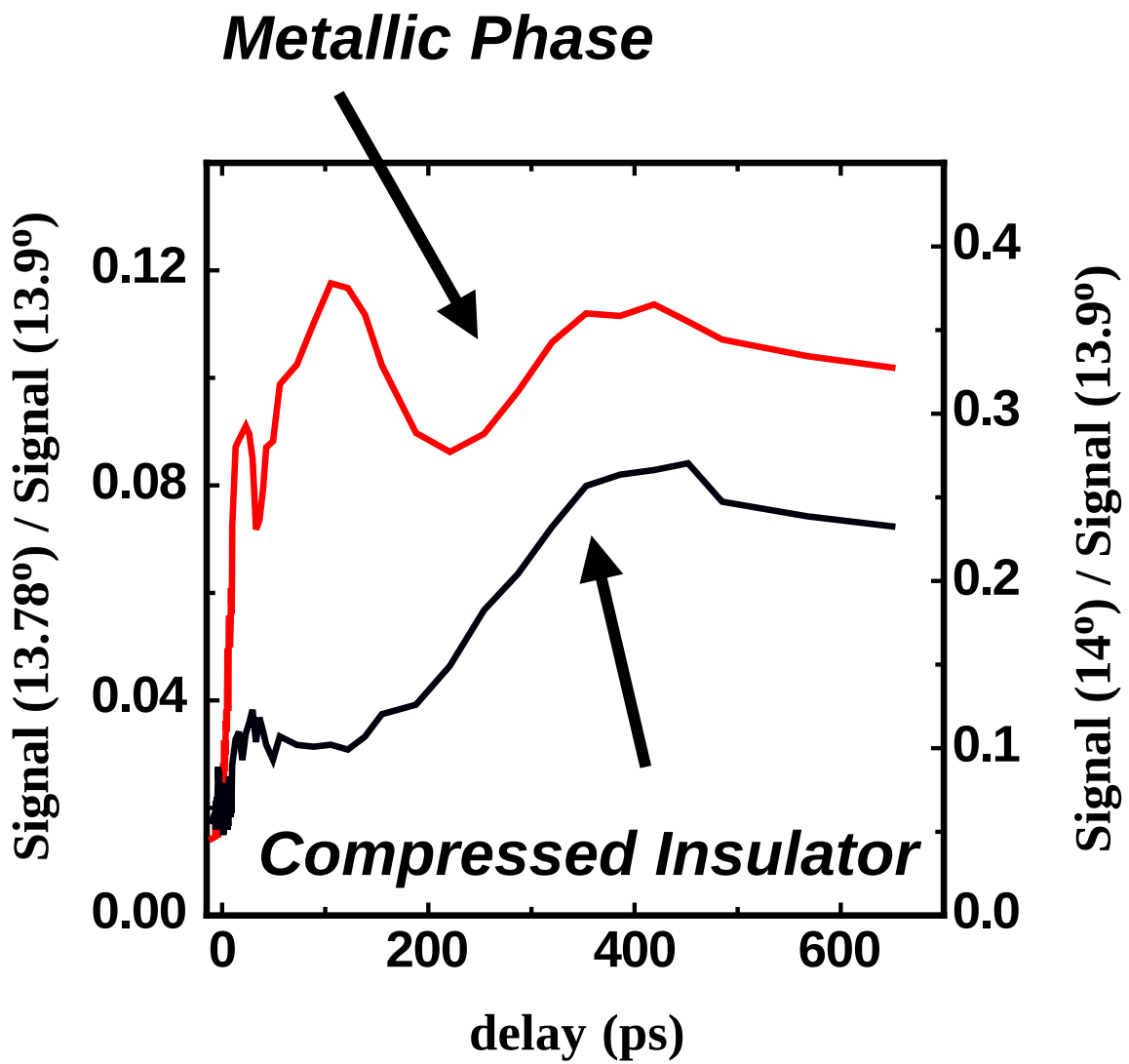
Compression of the Insulating phase



Compression of the insulating phase

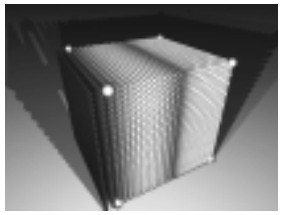


Compression of the insulating phase



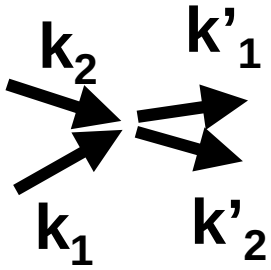
Summary and conclusion

Coherent lattice transport

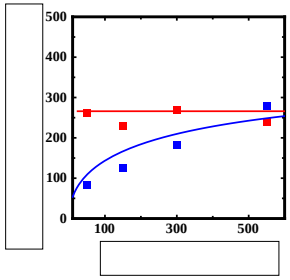


Lattice transport across buried interfaces

De-phasing of Acoustic phonons

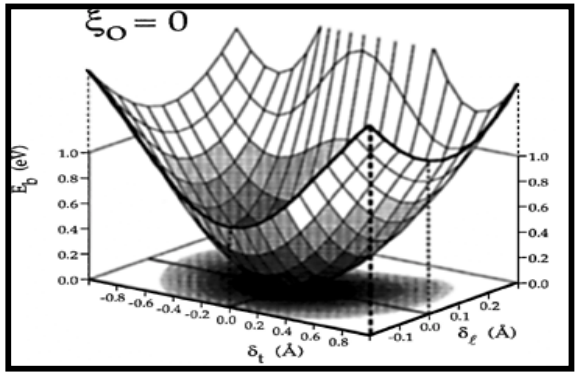


Heating depths and scaling laws for melting / ablation thresholds

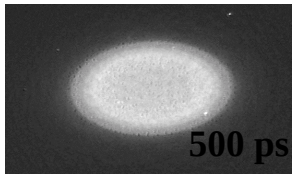


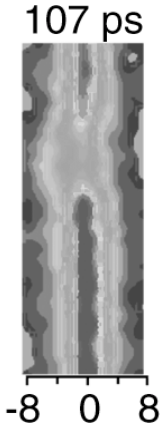
Conclusion

Thermal melting

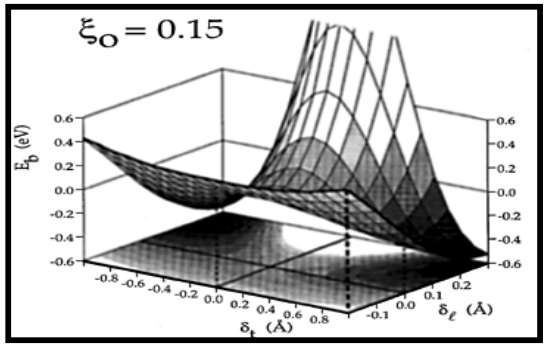


t > 100 ps

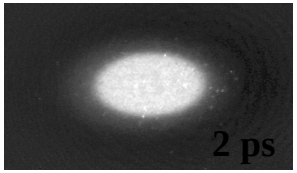


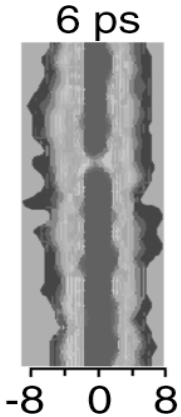


Non-thermal melting



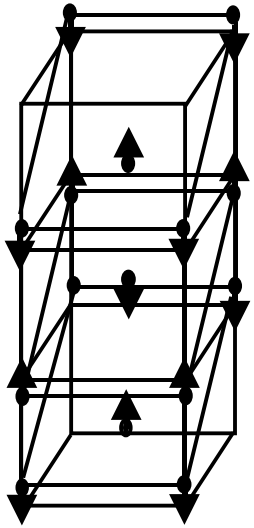
t ≈ 1 ps



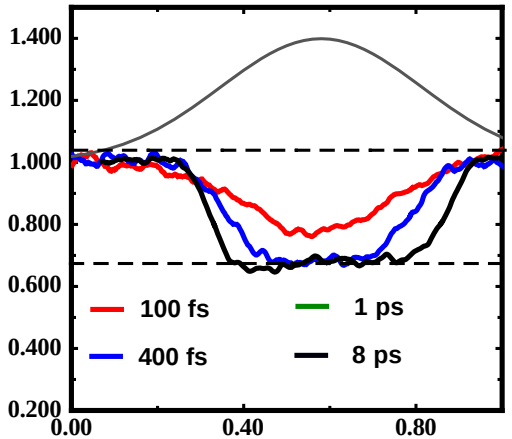
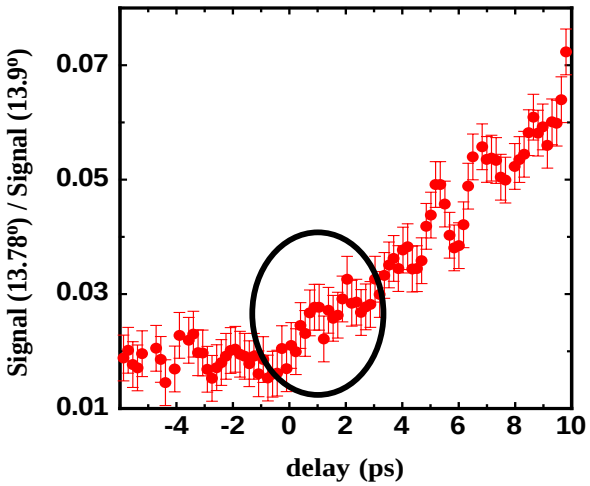


Conclusions

Solid-Solid phase transition in VO_2



Optical data



X-ray data