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Probing the disassembly of ultrafast laser heated gold using frequency domain interferometry

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Acknowledgements

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Optical measurements revealed a quasi-steady state in ultrafast laser excitation of gold

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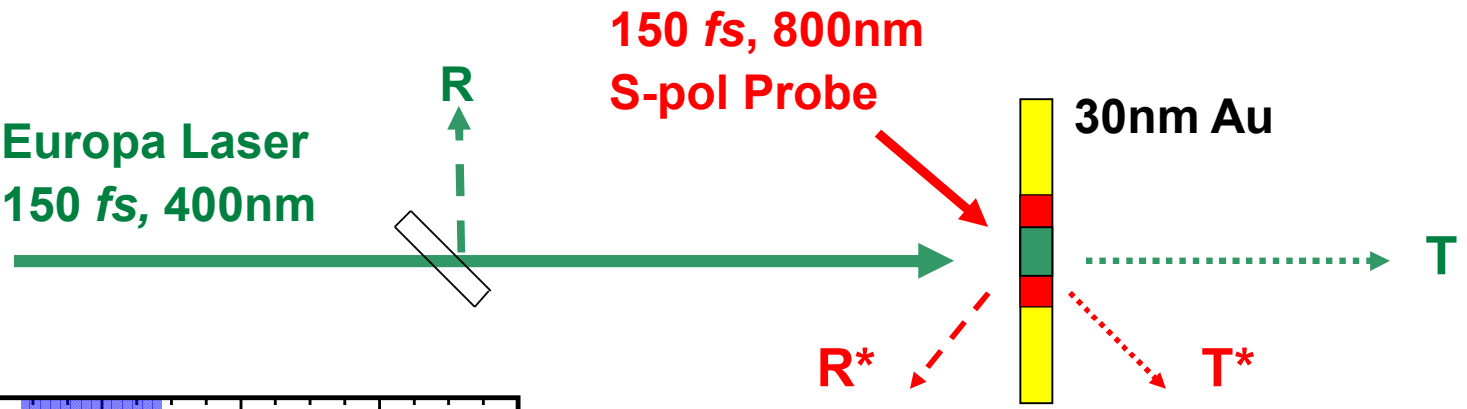
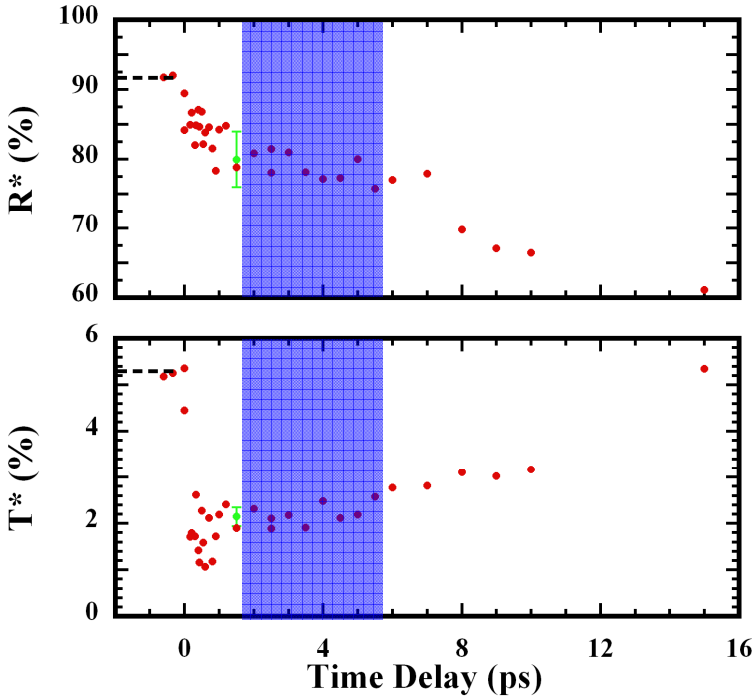
Widmann *et. al.*, PRL 92, 125002 (2004)





- Three distinct stages observed
  - –Initial transient : laser heating
  - –Quasi-steady state
  - –Second transient : hydro exp.

The quasi-steady state is an important finding

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- It ensures the applicability of the Idealized Slab Plasma (ISP) concept
  - Quasi-steady state signals the absence of significant hydrodynamic expansion
- It enables the viability of a much broader range of ps diagnostics for attaining single-state measurements

A critical question is the phase of the quasi-steady state

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- Calculations of transport properties require phase information, solid versus liquid, to determine the structure factor of the state

- The identity of the phase is also key to the understanding of non-equilibrium phase transitions induced by ultrafast excitation

This talk describes a first attempt to shed light on the quasi-steady state

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- Independent measurements corroborating the existence of the quasi-steady state
- Determination of the characteristic time for disassembly of the laser excited solid
- Identification of the phase of the quasi-steady state based on other findings

To monitor hydro expansion, we use the more sensitive P-pol R/T & S/P-pol FDI

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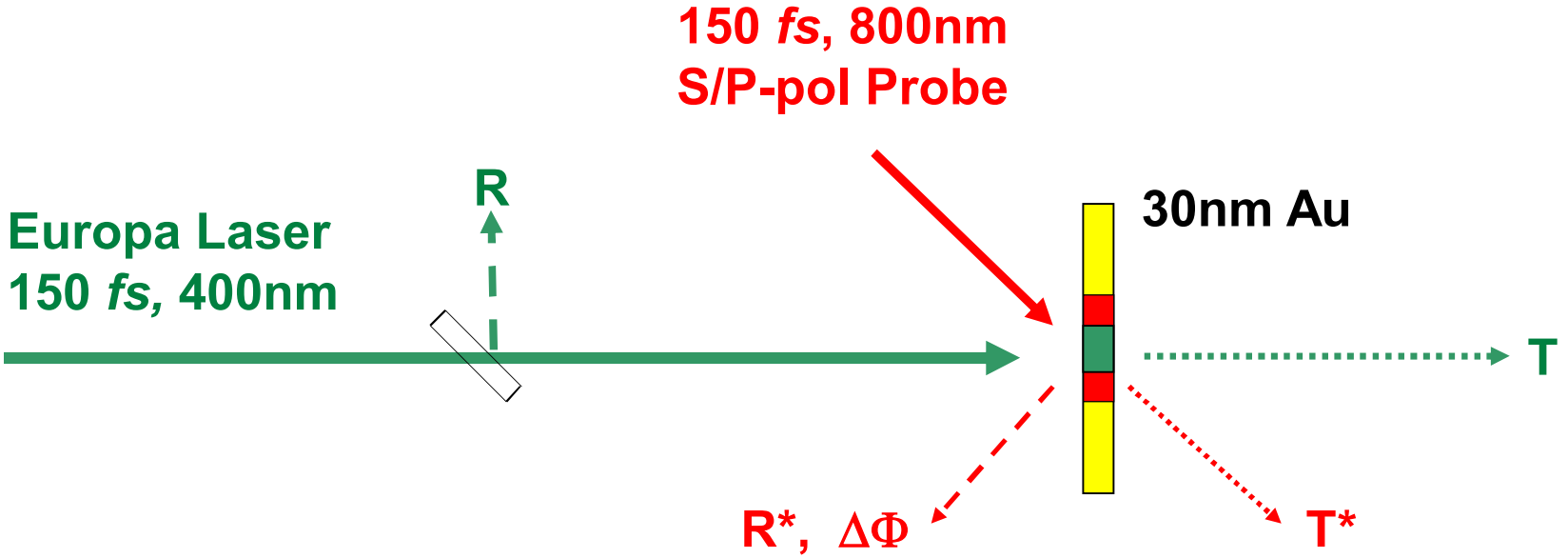

- **P-pol R/T** measurements are sensitive to **resonant absorption** in the critical density region of an expanding plasma
- **Frequency Domain Interferometry** monitors change in phase shift of a S/P-pol probe induced by **motion and/or changes in dielectric function** of heated foil

The FDI setup is a little more complex

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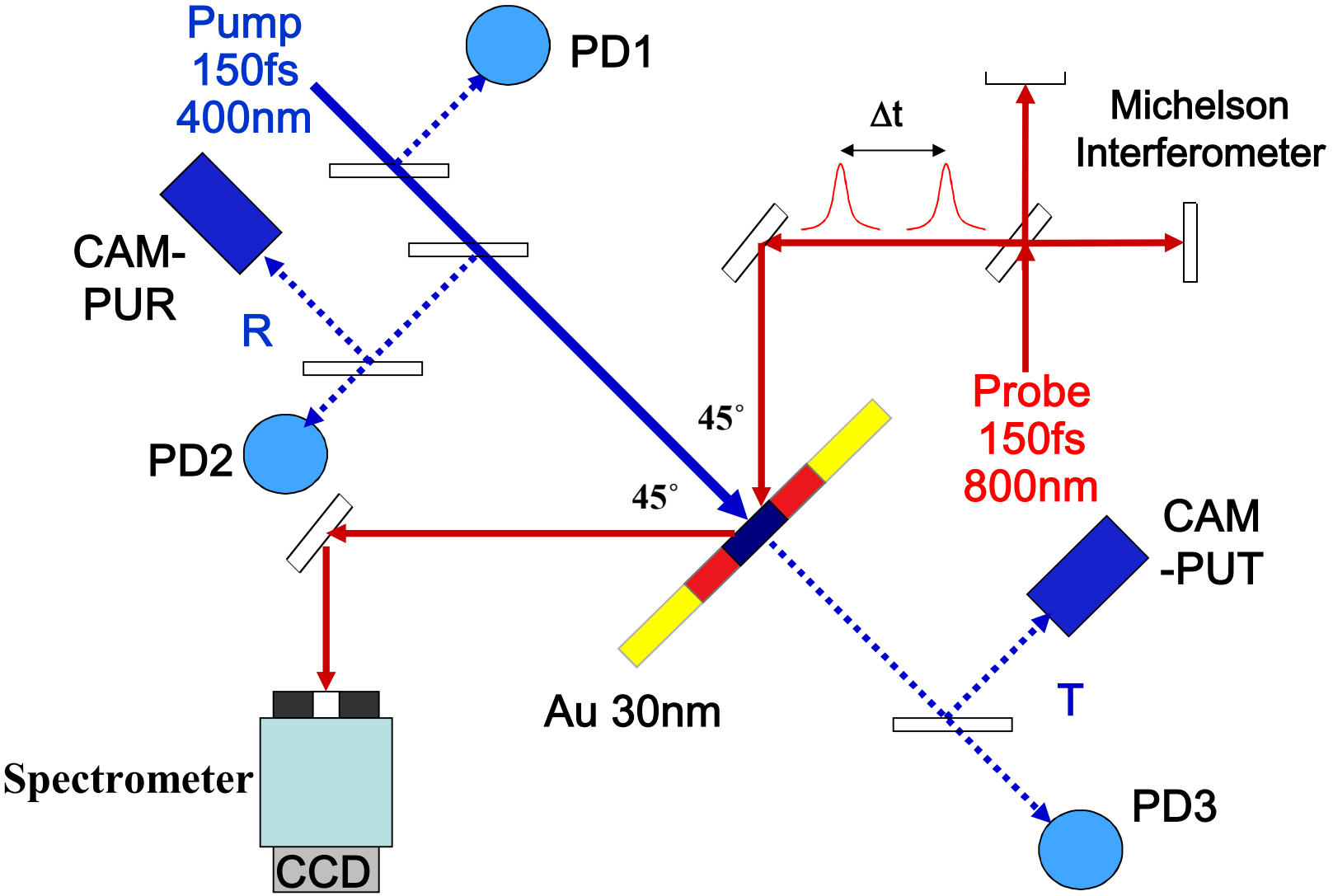

FDI able to detect small changes in phase shift (~10 mrads)

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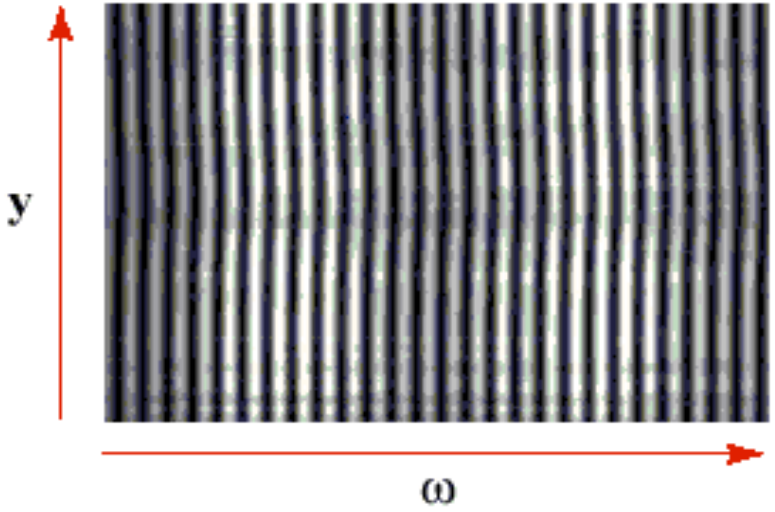

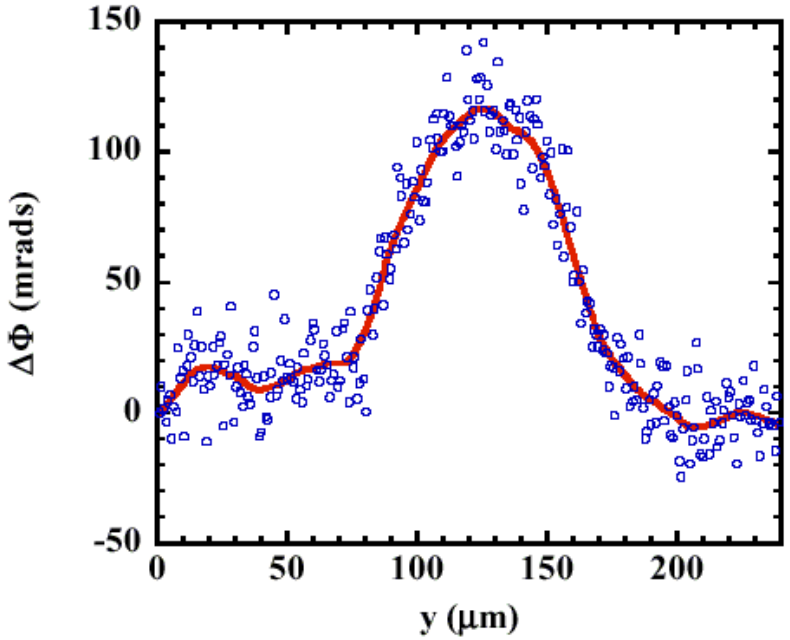

Quasi-steady state is confirmed in six different measurements

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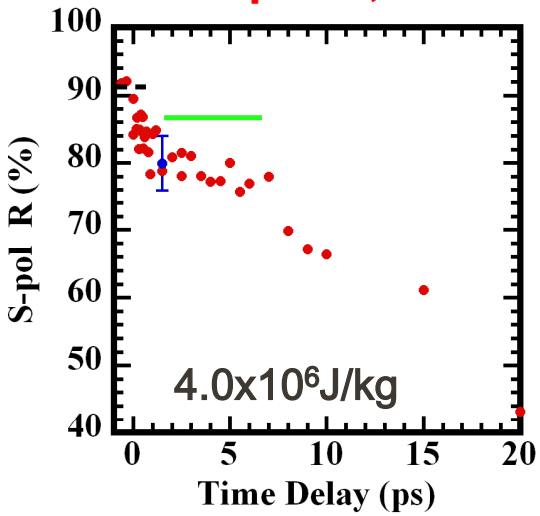
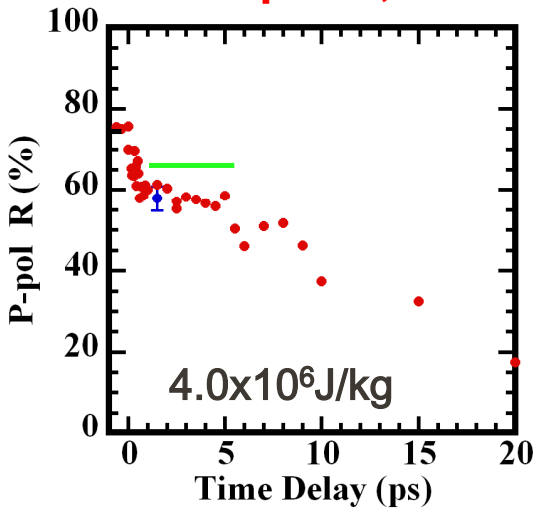
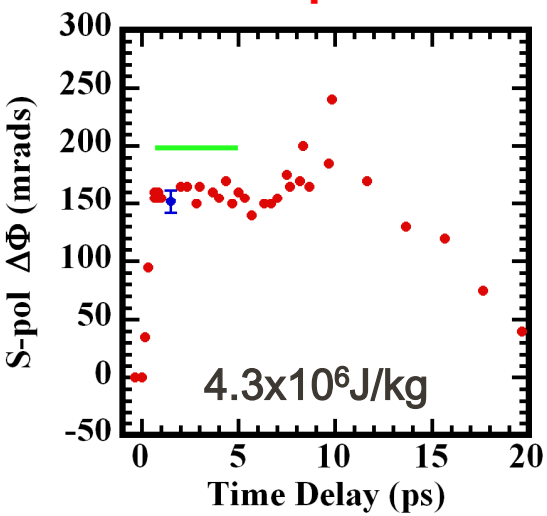
S-pol R, T

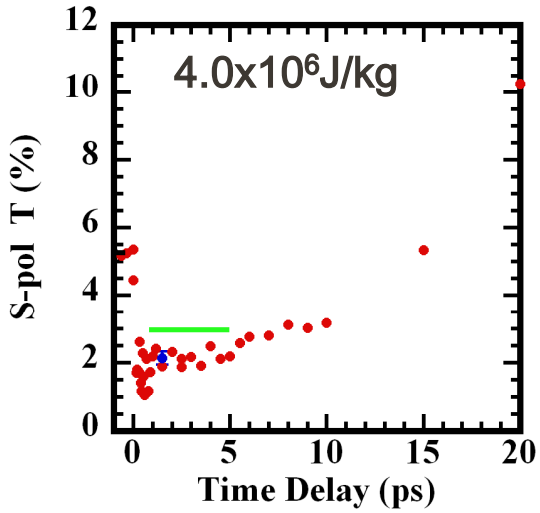


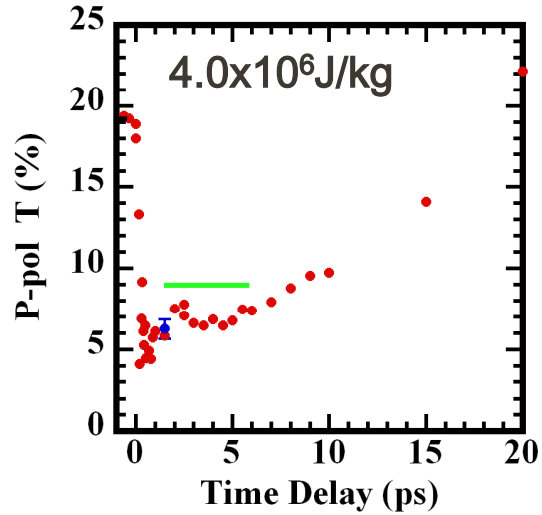
P-pol R, T

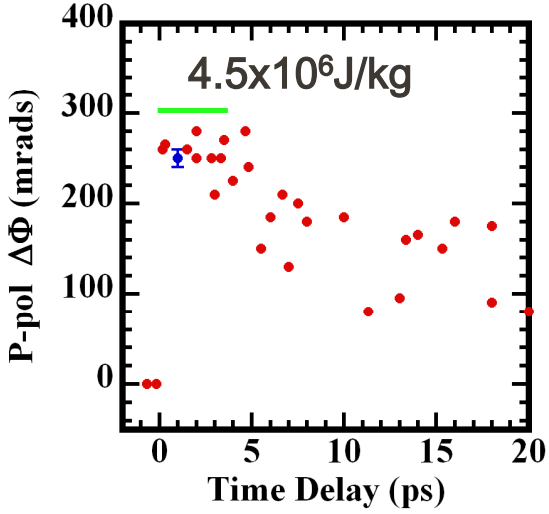


S/P-pol ΔΦ









To quantify the quasi-steady state duration, we use an extensive set of S-pol FDI data

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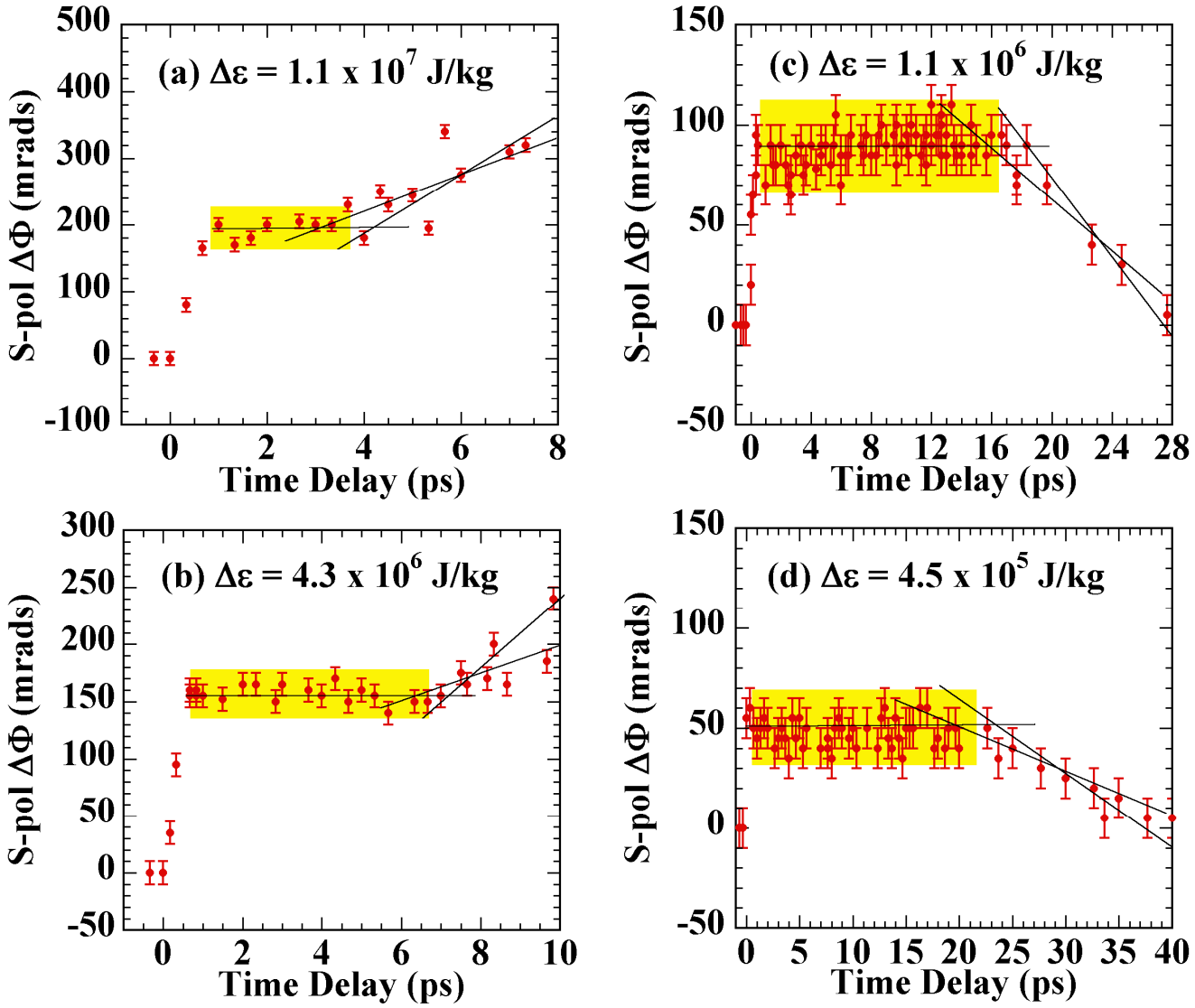

The quasi-steady state duration τ_{QSS} shows a clear dependence on excitation energy density

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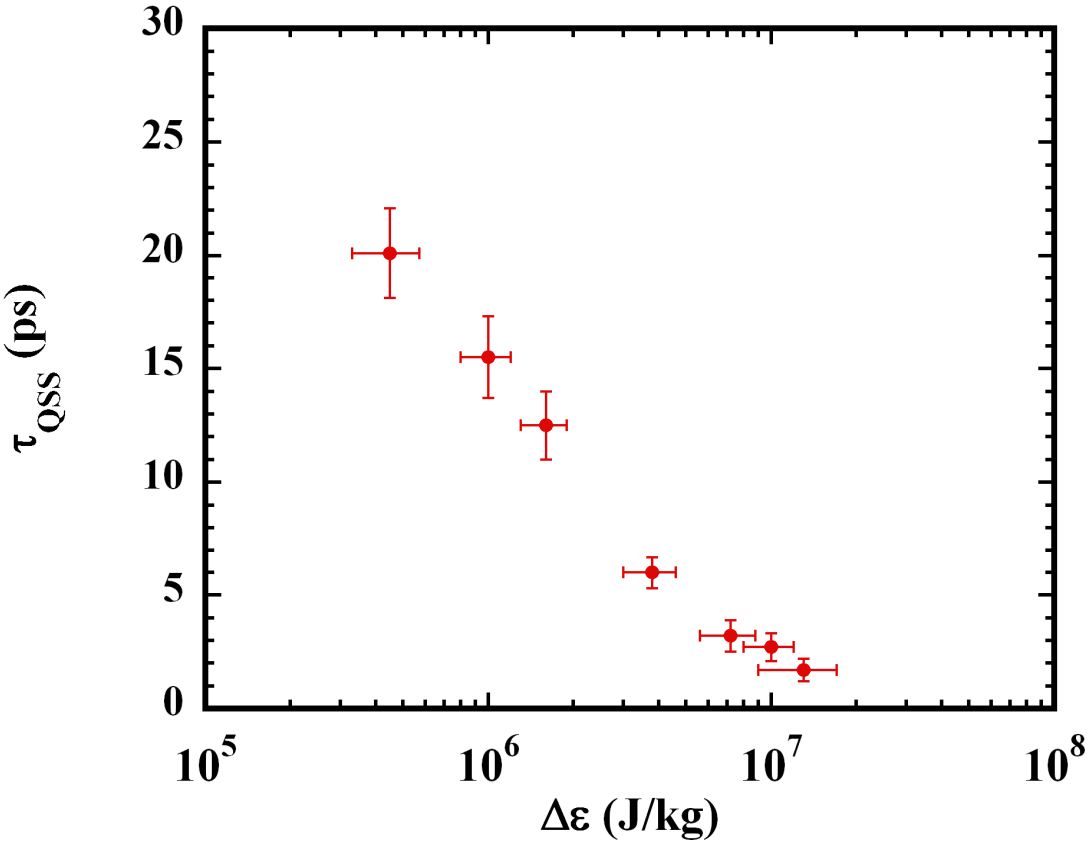
- The range of τ_{QSS} from 2-20 ps is substantial



Now, let us examine the dynamical processes occurring in ultrafast laser excitation

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- Pump excitation of d-electrons to the s/p band
- Thermalization gives rise to electron heating in the s/p band
- The heated electrons expand to form a sheath on the foil surface; the extent of the sheath is limited by space charge field
- Lattice heating is effected by electron-phonon coupling

To describe lattice heating, we use a modified Two-Temperature Model

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TTM: $$C_e(T_e)\frac{dT_e(t)}{dt} = -g\left[T_e(t) - \varepsilon_l(t)\frac{\rho_{Au}}{C_l}\right] + S(t)$$

$$\rho_{Au}\frac{d(\varepsilon_l(t))}{dt} = g\left[T_e(t) - \varepsilon_l(t)\frac{\rho_{Au}}{C_l}\right], \qquad \varepsilon_l(t) = \frac{C_l T_l(t)}{\rho_{Au}}$$

Electron-phonon coupling: $g = (2.2\pm0.3)\text{x}10^{16}$ W/m^3·K*

Heat capacities: $$C_e(T_e) = \frac{\partial U_e(T_e)}{\partial T_e}, \qquad C_l = 2.5\text{x}10^6 \text{ J/m}^3\text{·K}\dagger$$

Laser energy deposition: $$S(t) = \frac{\Delta\varepsilon\rho_{Au}}{\tau_P\sqrt{\pi}}\exp\left(-\frac{t^2}{\tau_P^2}\right)$$

*Hohlfeld *et al.* Chem. Phys. 251, 237 (2000)

†Maxmillian's Chemical and Physical Data, Maxmillian Press, London, 1992

We postulate that disassembly occurs at a critical lattice energy density ε_D

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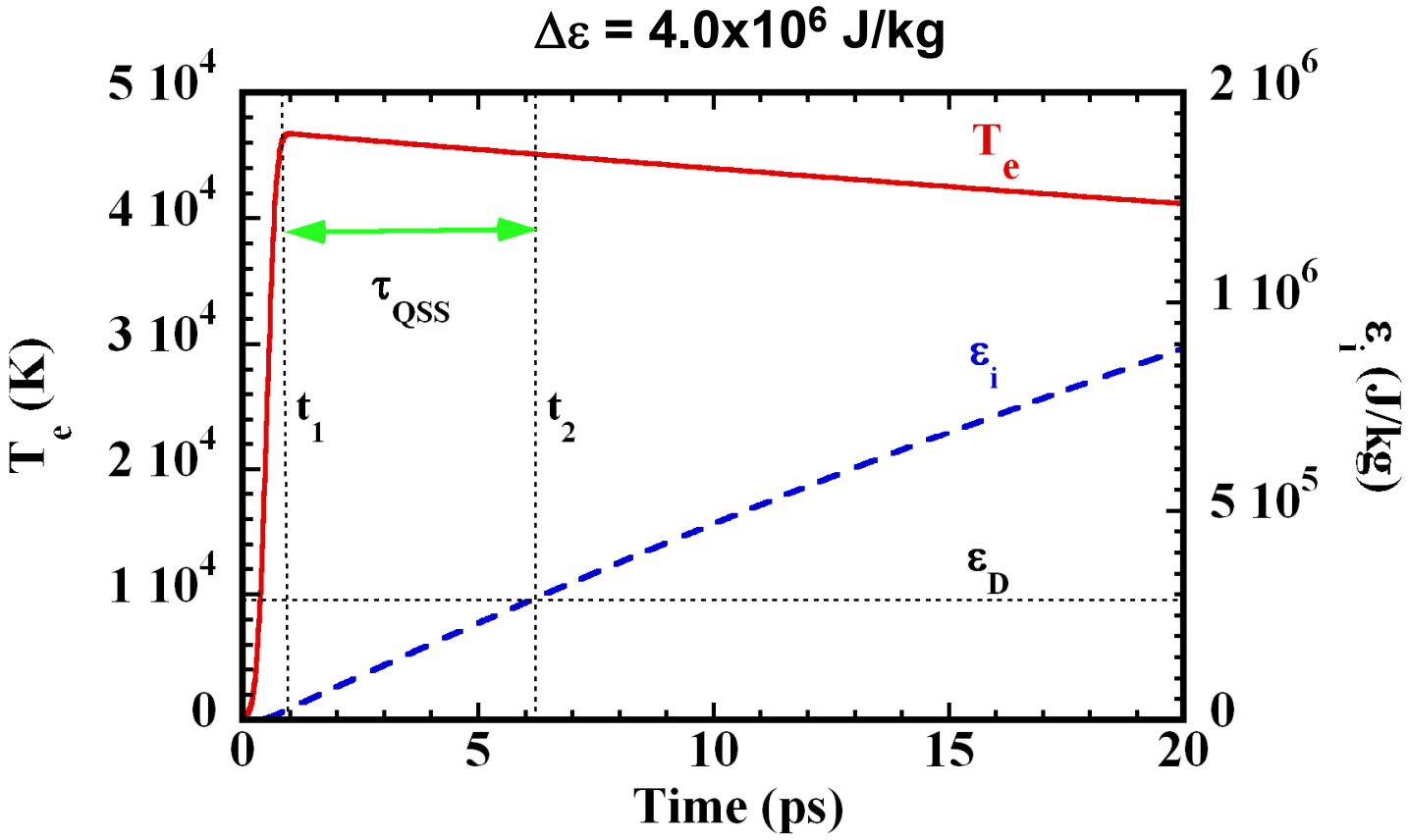
- Quasi-steady-state duration τ_{QSS} is determined by the value of ε_D that is taken to be constant, independent of heating rate (or $\Delta\varepsilon$)



The model appears to be consistent with observation

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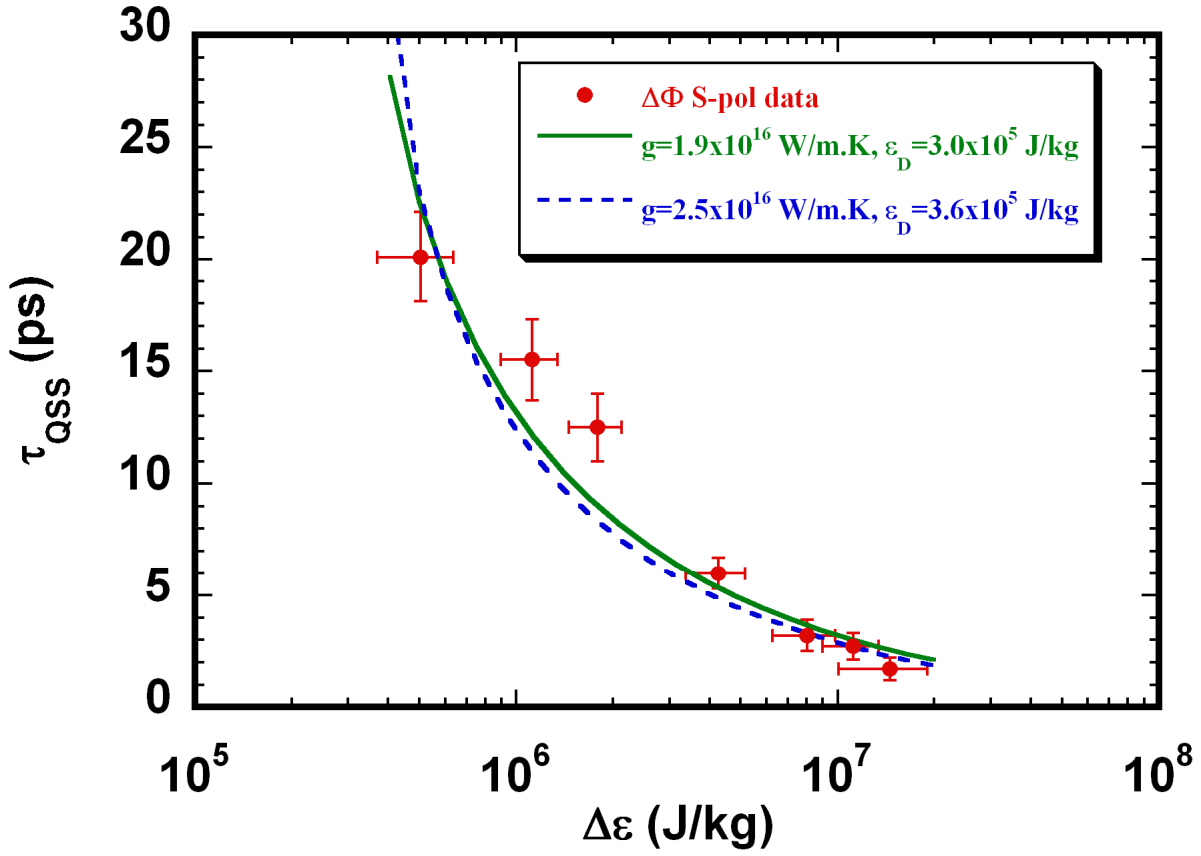
- Calculations with ε_D = (3.3±0.3)x10^5 J/kg show good agreement with data



To identify the phase of the quasi-steady state, we examine its dielectric function

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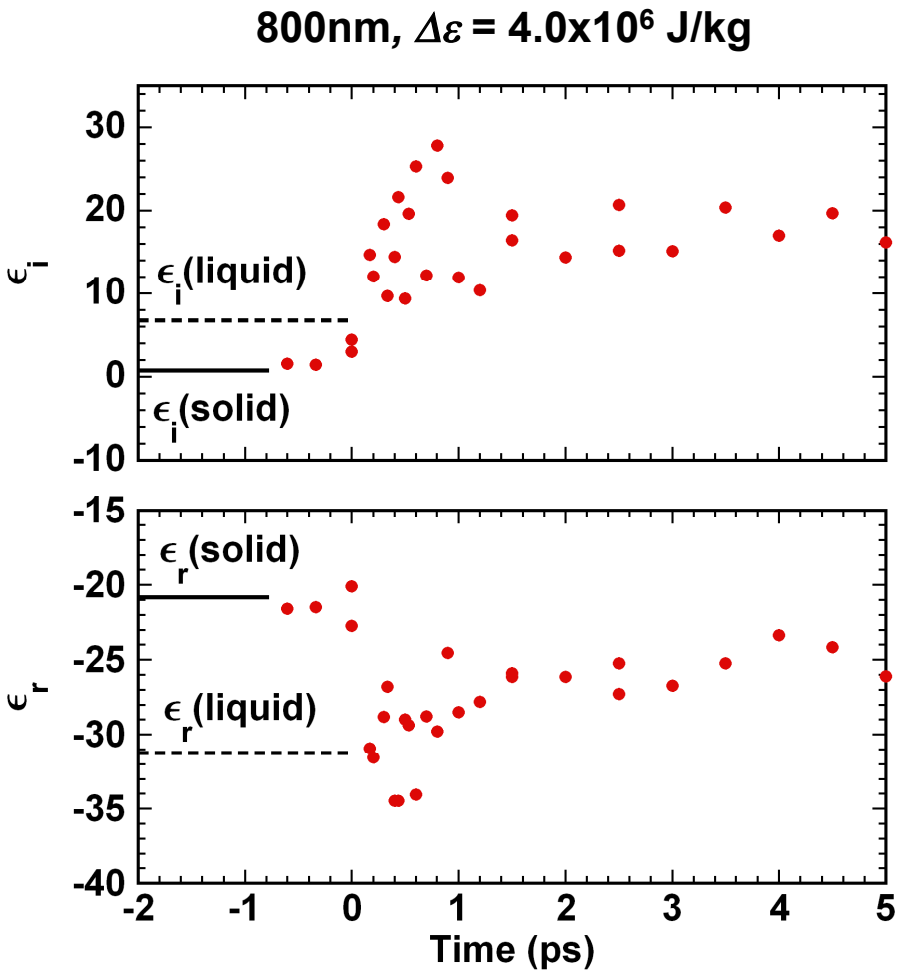

- Following the non-thermal melting interpretation of Guo *et. al.* [PRL, 84, 4493 (2000)], ϵ(t) suggests a solid-liquid transition within ~200fs
- The quasi-steady state would then be a liquid phase

Caveat: use of ∈ for an equilibrium liquid might be inappropriate

UED measurement on fs-laser heated Al has also indicated the possibility of a quasi-steady state liquid phase

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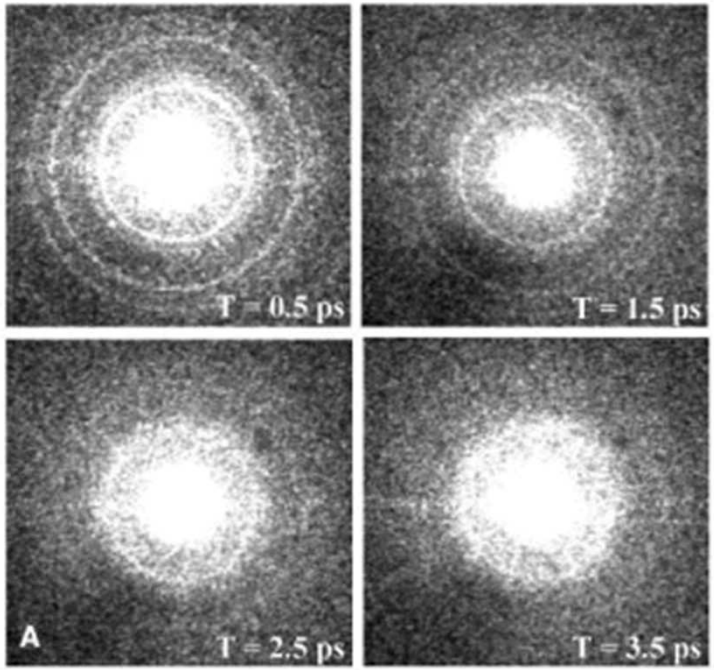
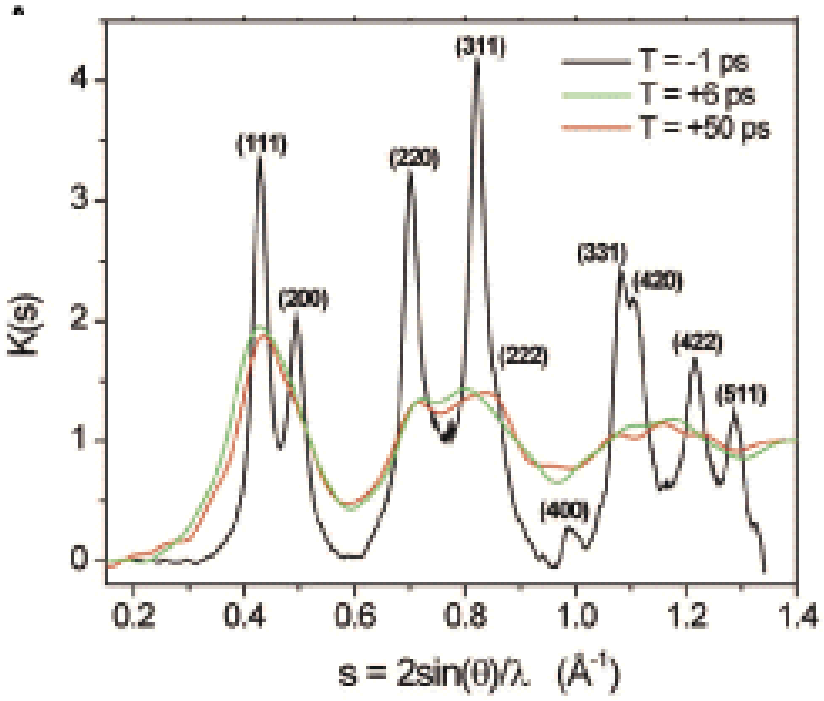
Siwick *et al.*, Science 302, 1382 (2002)

- Disordering to a liquid occurs in ~ 3ps
- Liquid structure appears unchanged to ~50ps





Caveat: measurement for Al at lower energy densities

Conclusions

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- Quasi-steady state in ultrafast laser excitation of gold has been confirmed in six different measurements
- The observed duration of the quasi-steady state can be described by a Two-Temperature Model with the postulate that disassembly occurs at a critical energy density of the lattice
- Following existing interpretations, the quasi-steady state can be taken to be a liquid phase

New measurements are needed for validation