Ultrafast (but Many-Body) Relaxation in a Low-Density Electron Glass

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Helgren, Armitage, Gruner PRL 2002

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Ultrafast (but Many-Body) Relaxation in a Low-Density Electron Glass

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We present a study of the relaxation dynamics of the photoexcited conductivity of the impurity states in the low-density electronic glass, phosphorous-doped silicon Si:P. Using subband gap optical pump-terahertz probe spectroscopy we find strongly temperature- and fluence-dependent glassy power-law relaxation occurring over subnanosecond time scales. Such behavior is in contrast to the much longer time scales found in higher electron density glassy systems. We also find evidence for both multiparticle relaxation mechanisms and/or coupling to electronic collective modes and a low temperature quantum relaxational regime.

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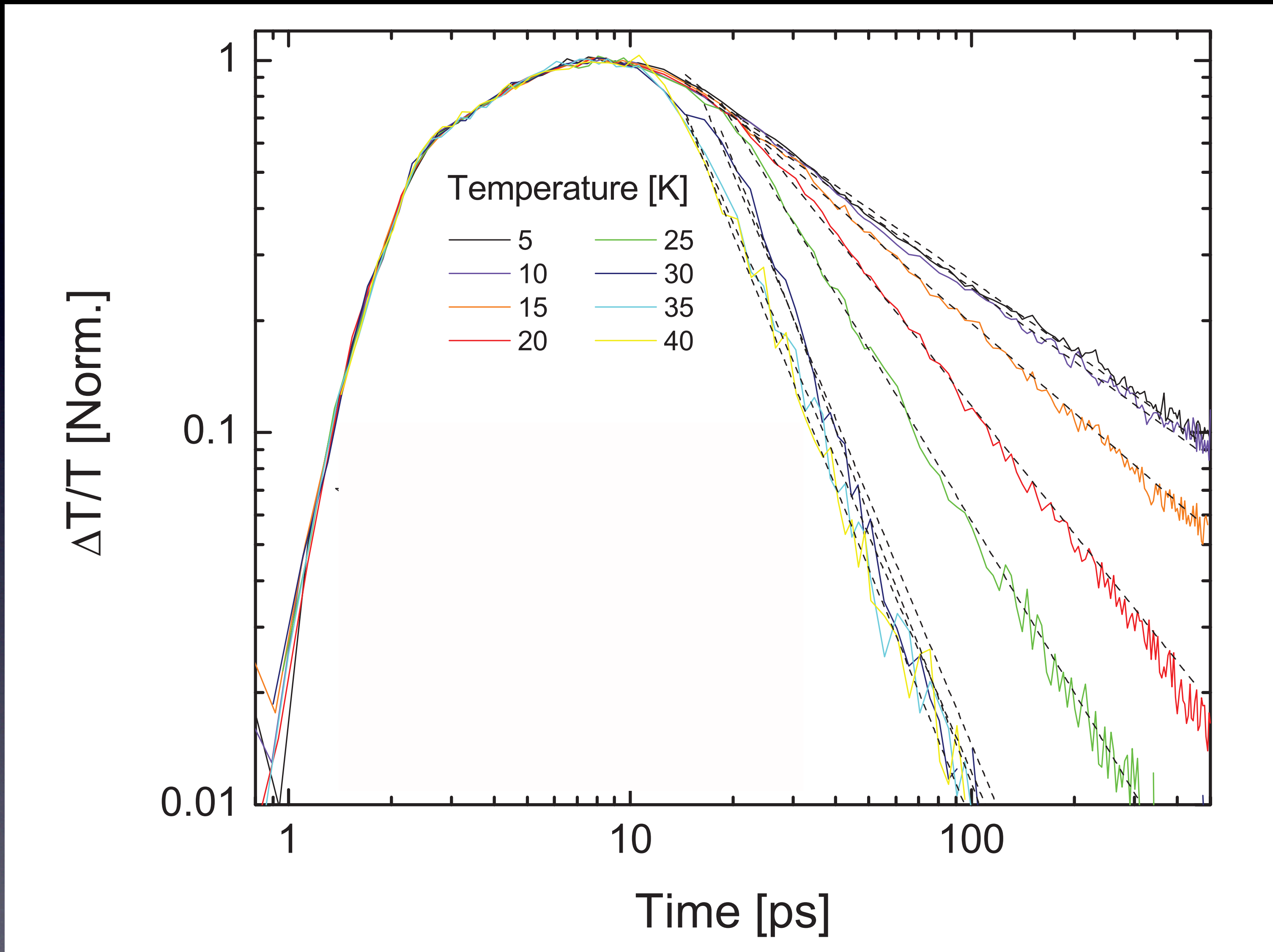
“Pump” Si:P with ultrafast laser pulse; Probe conductivity with THz pulse

After laser pulse charge decays quickly to impurity band...

Then “slow” *non-exponential* relaxation (100’s of ps);
“ultrafast” but slower than microscopic scales

Many-body relaxation? Connection to high density systems?

Transmission vs. pump/probe delay

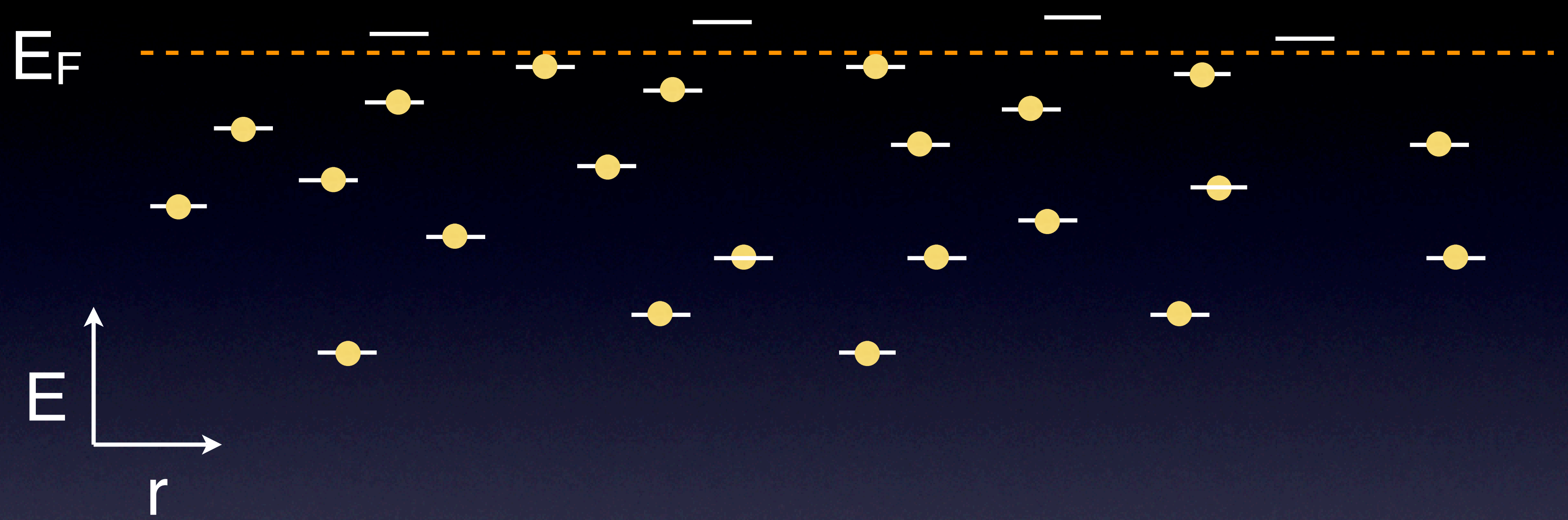


Glasses

Low-lying metastable states

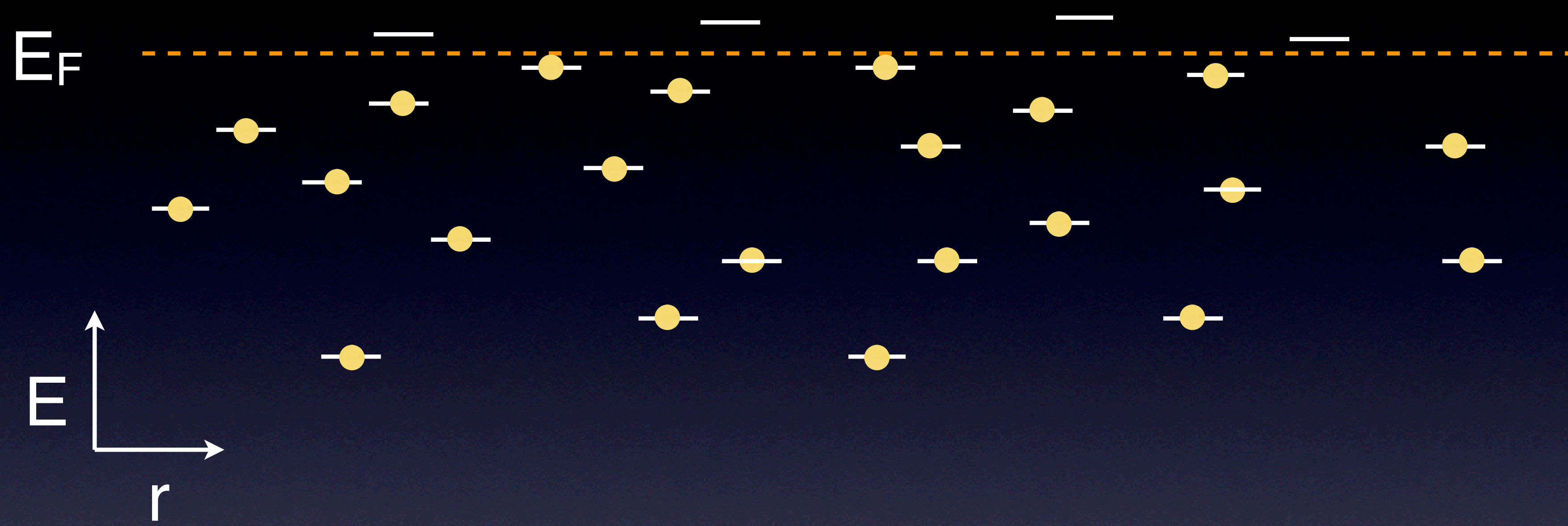
Disorder		Slow relaxation
Interactions	→	Non-exponential decay
Frustration		Aging
		Memory Effects

Electron Glasses



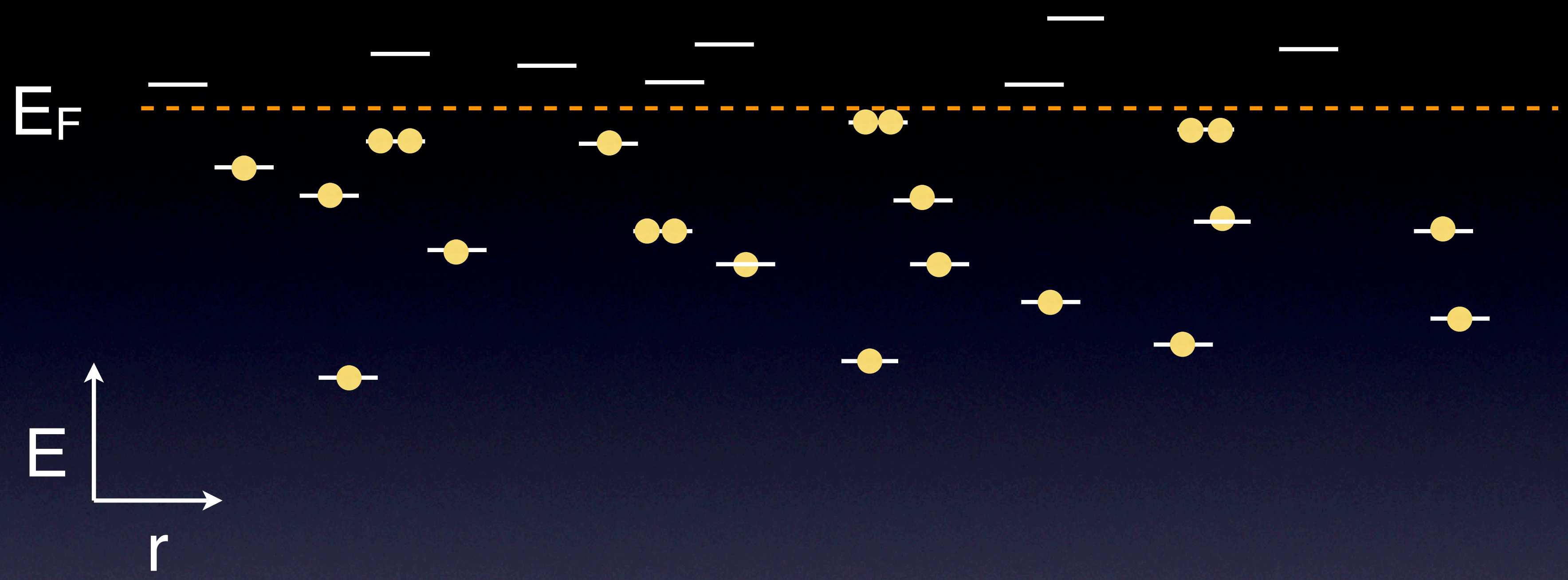
Ensemble of orbitals localized in real space

Electron Glasses



What about interactions?

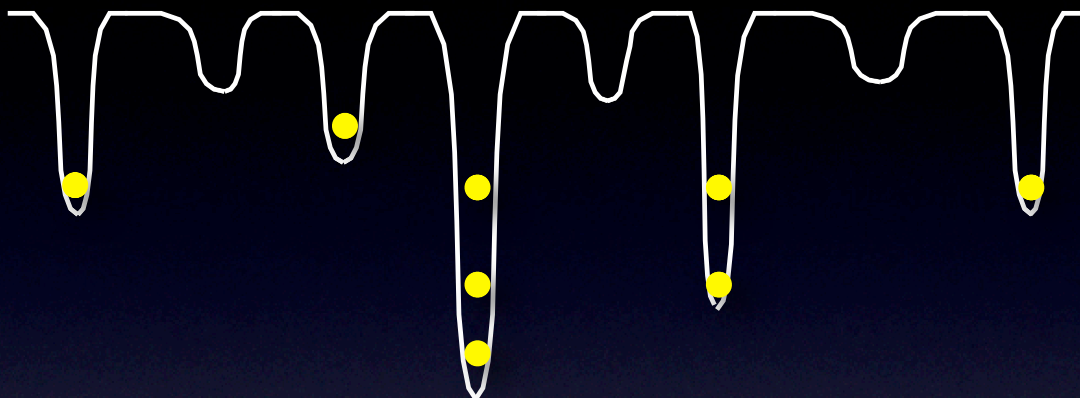
Electron Glasses



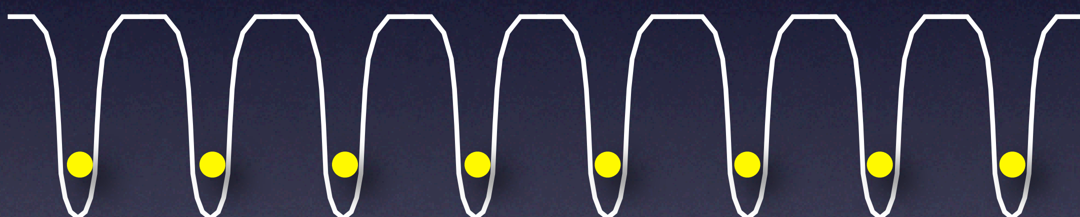
Long-range *1/r* Coulomb interaction; no screening

Energy levels are not just a function of local disorder and site occupations; energy is a strong function of other occupations

Interactions + Disorder = Frustration!



Disorder → Nonuniform Density



Coulomb Interactions → Uniform Density

(variable range hopping transport (Mott and/or Efros Shklovskii); Coulomb gap, ferromagnetism)

What about the “glass” in electron glass?

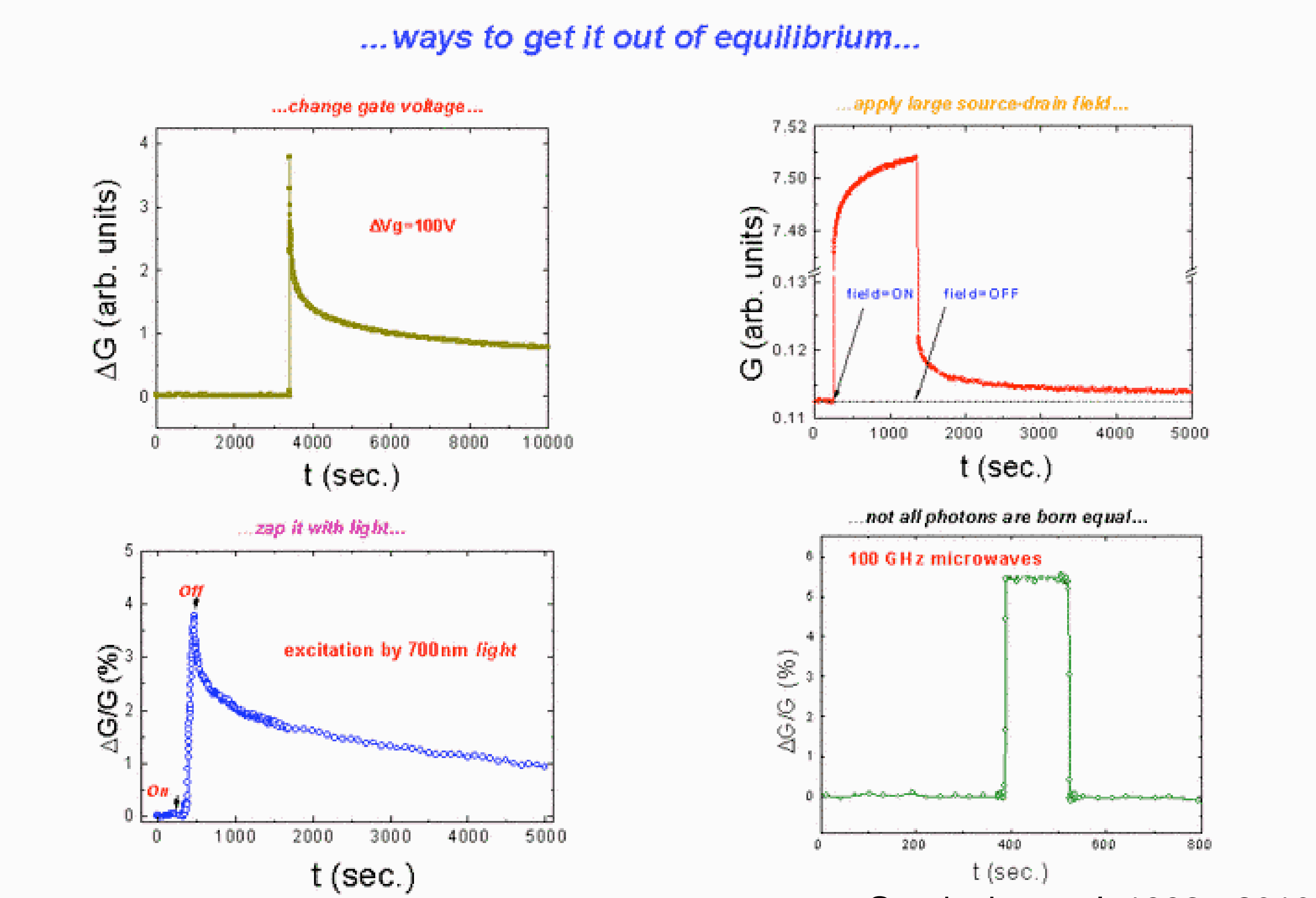
Many metastable quasi-ground states with large potential barriers between them...

Hard to reach true ground state; single particle hops eventually increase energy...

Multi-particle hops are required to lower energy, but rare and inefficient...

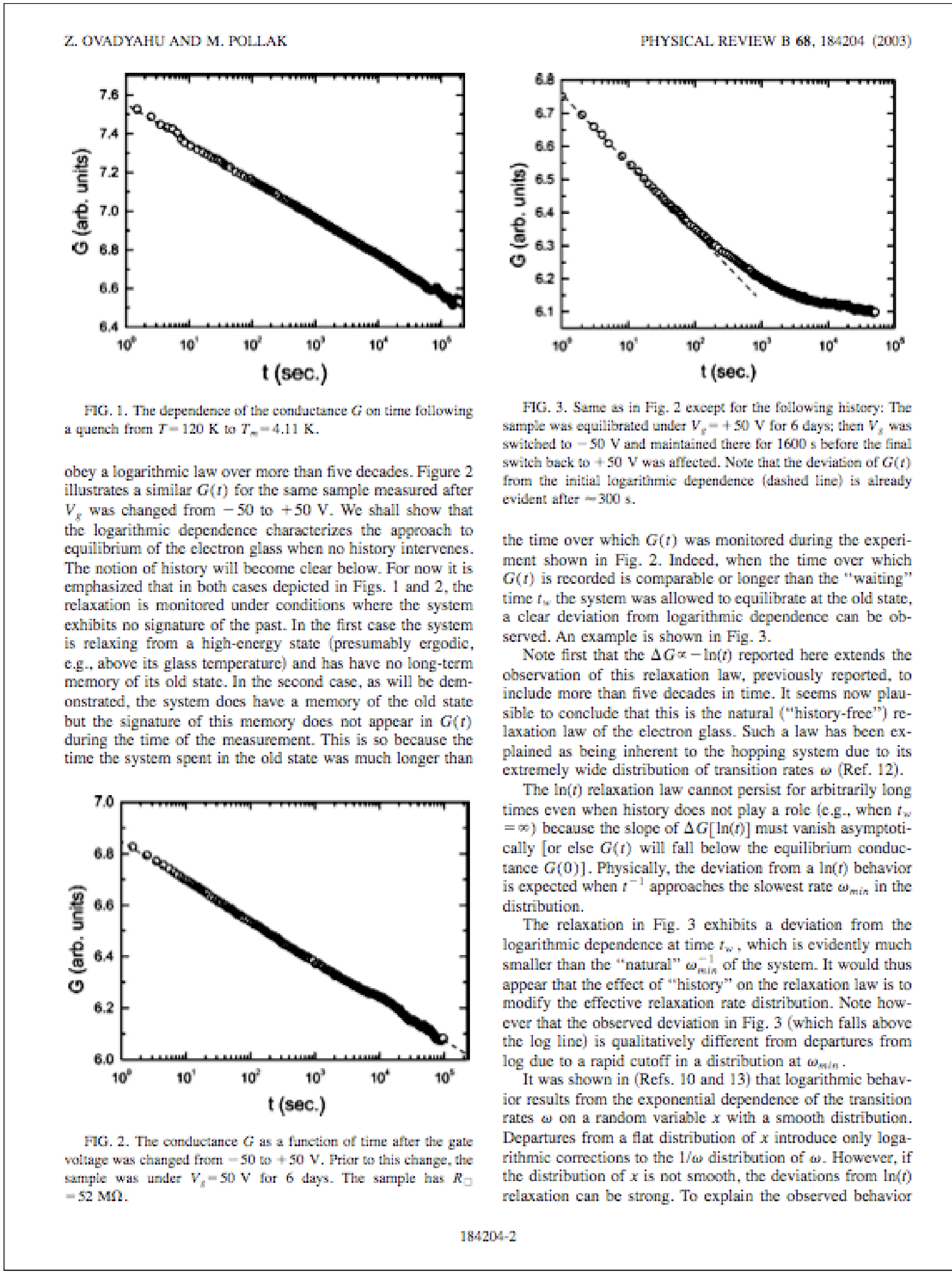
Lots of work on equilibrium properties like hopping conductivity (variable range), tunneling etc. but until recently much less showing explicitly glassy phenomena (relaxation)

Relaxation in InO_x films (similar in granular metals)



Ovadyahu et al. 1993 - 2010

5000 seconds ~ 1.4 hours



Z. OVADYAHU AND M. POLLAK PHYSICAL REVIEW B **68**, 184204 (2003)

FIG. 1. The dependence of the conductance G on time following a quench from $T=120$ K to $T_m=4.11$ K.

obey a logarithmic law over more than five decades. Figure 2 illustrates a similar $G(t)$ for the same sample measured after V_g was changed from -50 to $+50$ V. We shall show that the logarithmic dependence characterizes the approach to equilibrium of the electron glass when no history intervenes. The notion of history will become clear below. For now it is emphasized that in both cases depicted in Figs. 1 and 2, the relaxation is monitored under conditions where the system exhibits no signature of the past. In the first case the system is relaxing from a high-energy state (presumably ergodic, e.g., above its glass temperature) and has have no long-term memory of its old state. In the second case, as will be demonstrated, the system does have a memory of the old state but the signature of this memory does not appear in $G(t)$ during the time of the measurement. This is so because the time the system spent in the old state was much longer than

FIG. 2. The conductance G as a function of time after the gate voltage was changed from -50 to $+50$ V. Prior to this change, the sample was under $V_g=50$ V for 6 days. The sample has $R_\square=52$ MΩ.

FIG. 3. Same as in Fig. 2 except for the following history: The sample was equilibrated under $V_g=+50$ V for 6 days; then V_g was switched to -50 V and maintained there for 1600 s before the final switch back to $+50$ V was affected. Note that the deviation of $G(t)$ from the initial logarithmic dependence (dashed line) is already evident after ≈ 300 s.

the time over which $G(t)$ was monitored during the experiment shown in Fig. 2. Indeed, when the time over which $G(t)$ is recorded is comparable or longer than the "waiting" time t_w the system was allowed to equilibrate at the old state, a clear deviation from logarithmic dependence can be observed. An example is shown in Fig. 3.

Note first that the $\Delta G \propto -\ln(t)$ reported here extends the observation of this relaxation law, previously reported, to include more than five decades in time. It seems now plausible to conclude that this is the natural ("history-free") relaxation law of the electron glass. Such a law has been explained as being inherent to the hopping system due to its extremely wide distribution of transition rates ω (Ref. 12).

The $\ln(t)$ relaxation law cannot persist for arbitrarily long times even when history does not play a role (e.g., when $t_w=\infty$) because the slope of $\Delta G[\ln(t)]$ must vanish asymptotically [or else $G(t)$ will fall below the equilibrium conductance $G(0)$]. Physically, the deviation from a $\ln(t)$ behavior is expected when t^{-1} approaches the slowest rate ω_{min} in the distribution.

The relaxation in Fig. 3 exhibits a deviation from the logarithmic dependence at time t_w, which is evidently much smaller than the "natural" ω_{min}^{-1} of the system. It would thus appear that the effect of "history" on the relaxation law is to modify the effective relaxation rate distribution. Note however that the observed deviation in Fig. 3 (which falls above the log line) is qualitatively different from departures from log due to a rapid cutoff in a distribution at ω_{min}.

It was shown in (Refs. 10 and 13) that logarithmic behavior results from the exponential dependence of the transition rates ω on a random variable x with a smooth distribution. Departures from a flat distribution of x introduce only logarithmic corrections to the $1/\omega$ distribution of ω. However, if the distribution of x is not smooth, the deviations from $\ln(t)$ relaxation can be strong. To explain the observed behavior

184204-2

"History free" relaxation is logarithmic

10^5 seconds ~ 27 hours

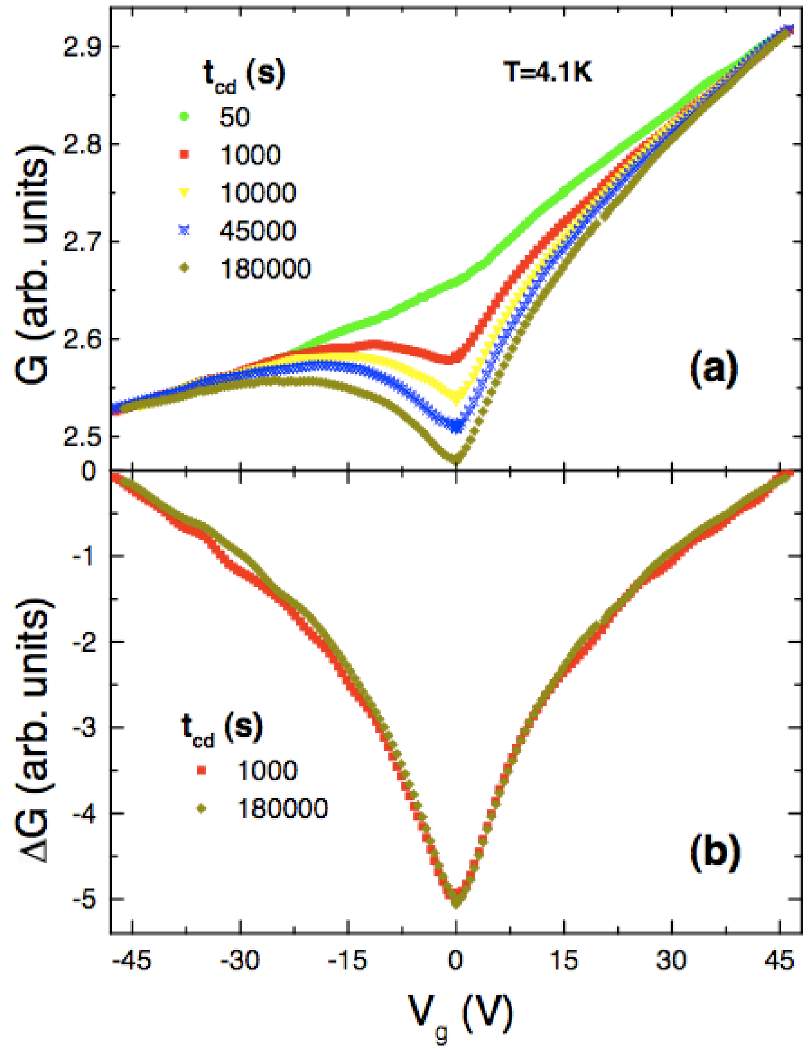


FIG. 5: (a) Field effect sweeps taken at several times since cool-down t_{cd} showing the MD slow evolution. Sample is In_2O_{3-x} film with $R = 49$ MΩ. (b) Shows the collapse of the MD shape for the data for the sweeps taken at $t_{cd} = 10^3$ seconds and $t_{cd} = 180,000$ seconds (after subtracting the equilibrium field effect and expanding the $t_{cd} = 10^3$ seconds MD data by a constant factor).

Phenomenologically consequence of "Memory Dip" effect

What controls the relaxation times?

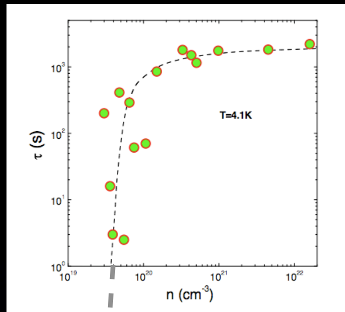


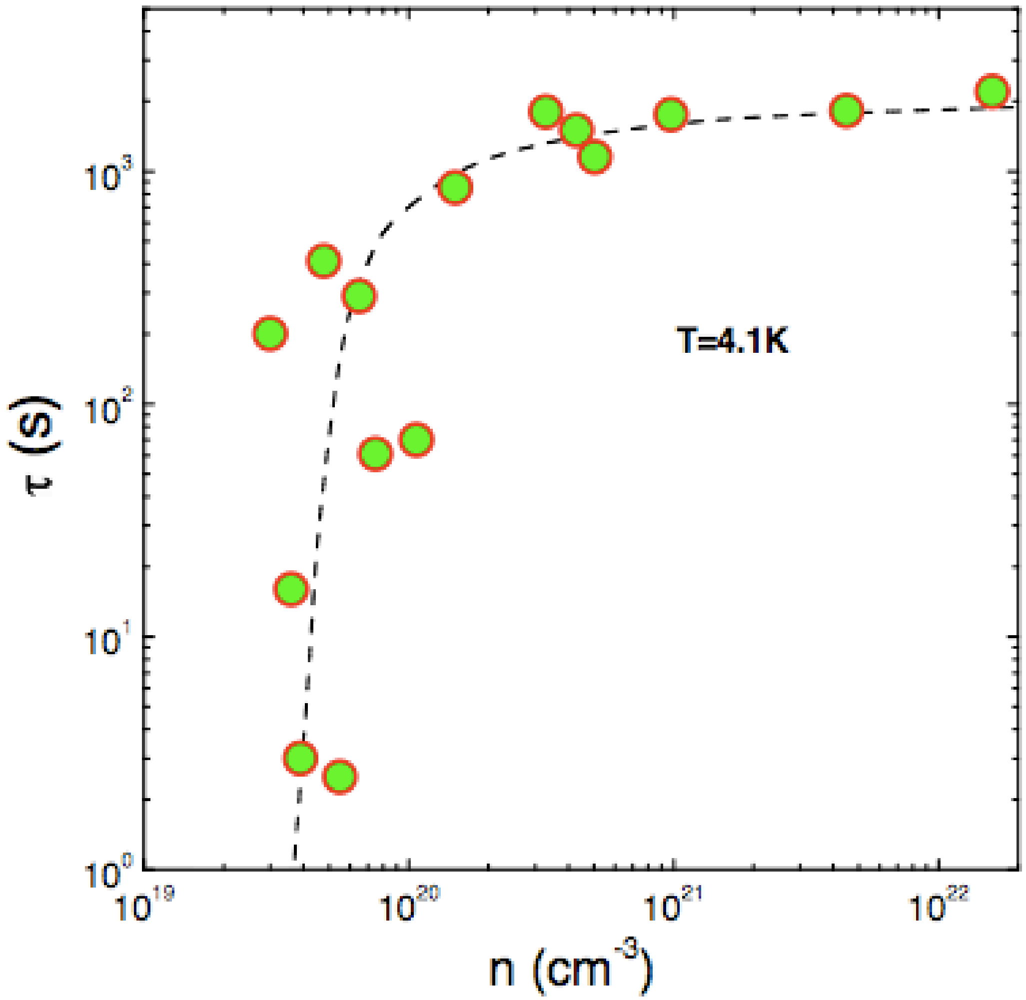
FIG. 3: Typical relaxation time τ, as function of the carrier concentration n for a series of In_xO samples (data are based on the two-dip experiment, see [17] and [28] for fuller details and interpretation). Note the sharp drop of τ for $n \lesssim 10^{20}$ cm^{-3}.

Relaxation times a function of density?

Connection to orthogonality of quasi-groundstates? Larger at high density?

What about very low densities?

$\Leftarrow$ << 10^{-15} sec @ 10^{18} charges/cm^3 ?



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Relaxation times a function of density?

Connection to orthogonality of quasi-groundstates? Larger at high density?

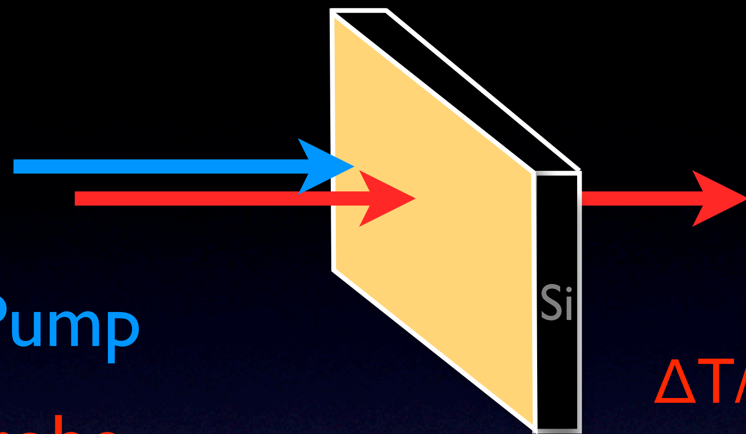
What about very low densities?

Semiconductors?

Considerations for fast measurements...

ω_{Nyq} or $\omega_{meas} > 1/\tau_{decay}$

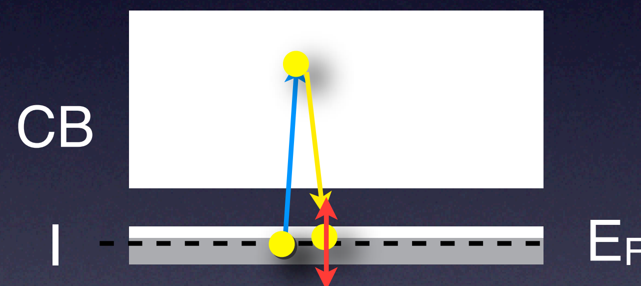
Optical Pump - THz Probe



0.8 eV Optical Pump

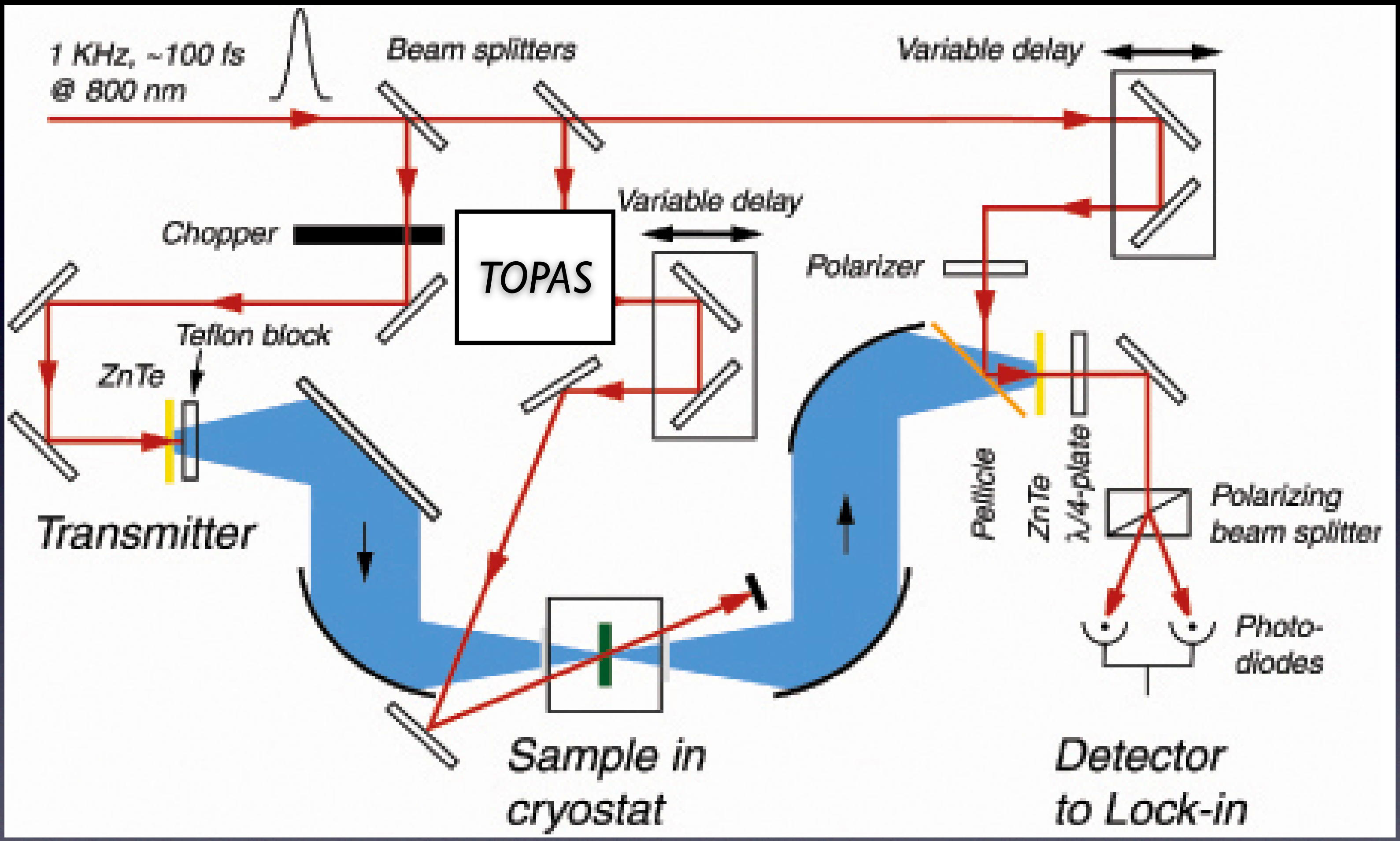
2.5 meV THz probe

$\Delta T/T \sim -\Delta\sigma/\sigma$

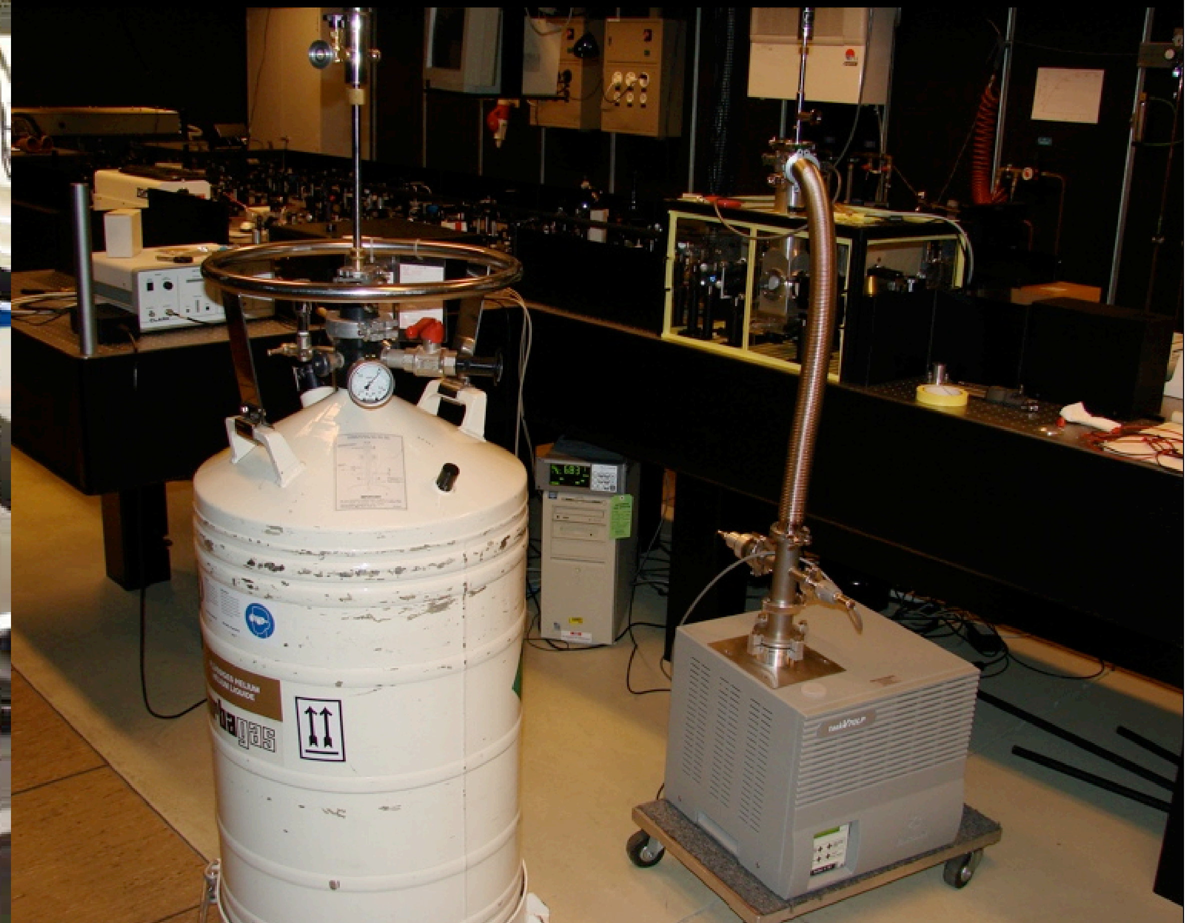


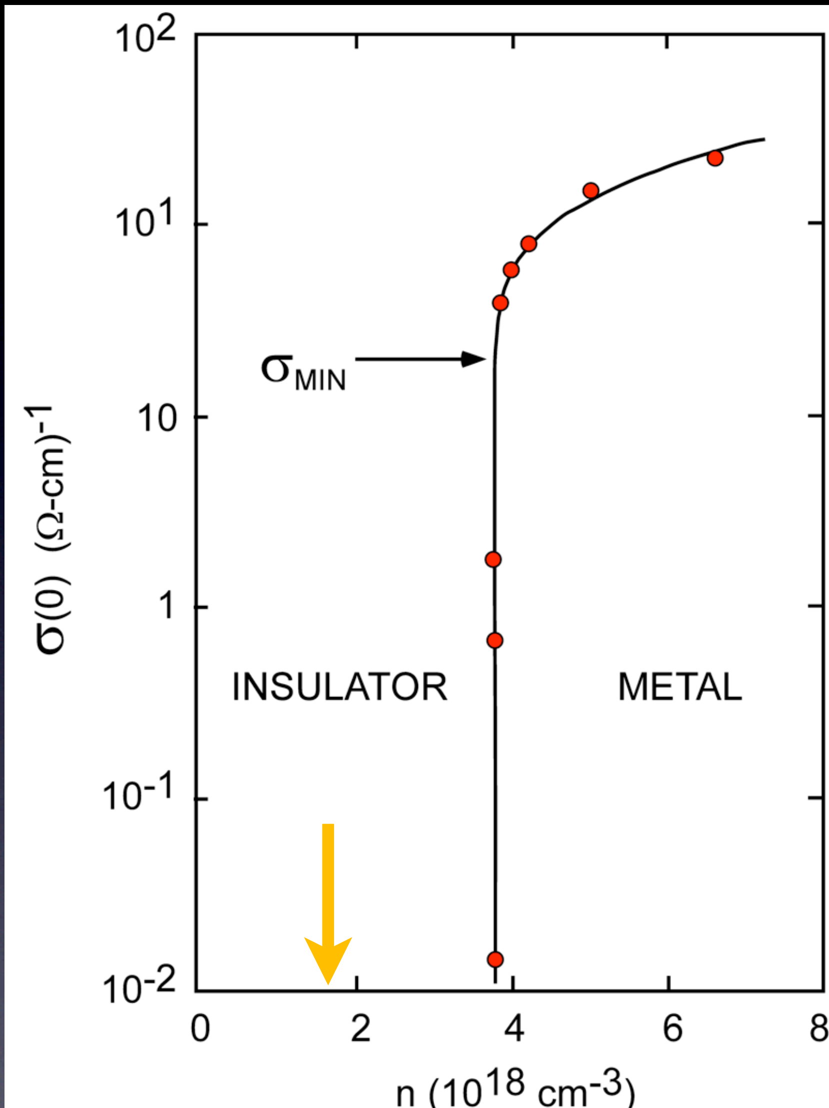
4.2 - 424 μJ/cm² Pump Fluence → $3.2 \times 10^{15}/cm^3$ - $3.2 \times 10^{17}/cm^3$ Excited Charge Density

Optical Pump - THz Probe



Optical Pump - THz Probe





Rosenbaum et al. PRL 1983

Lightly doped Si

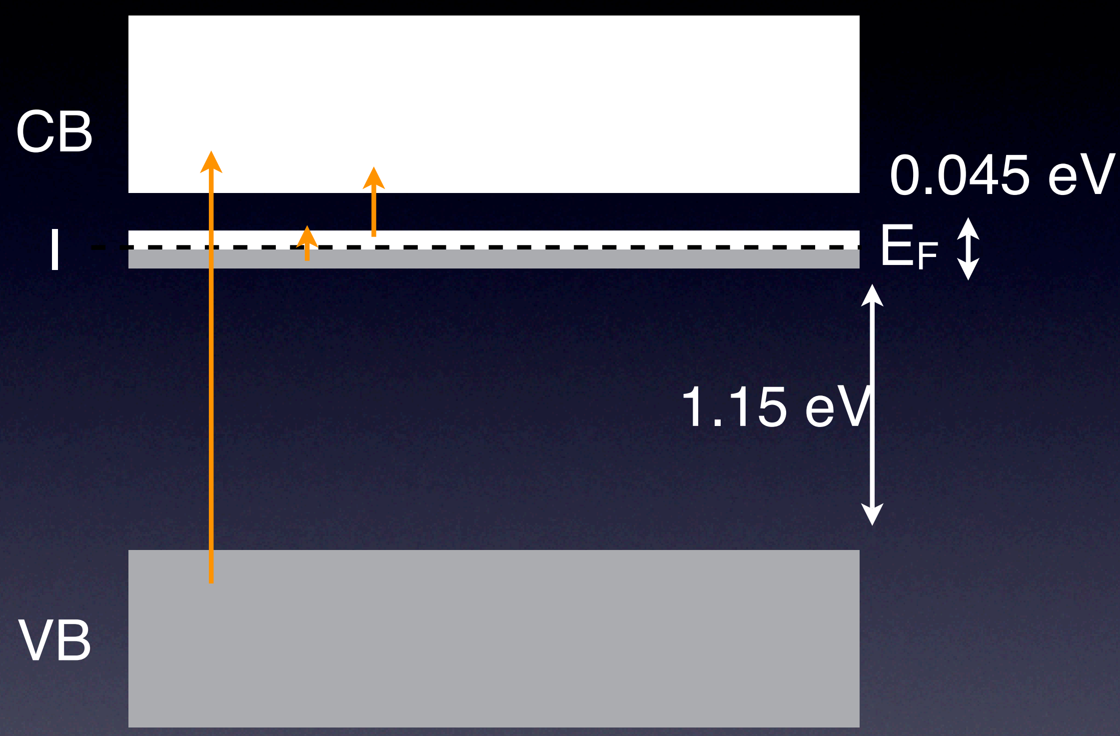
Si:P @ 39% of n_c

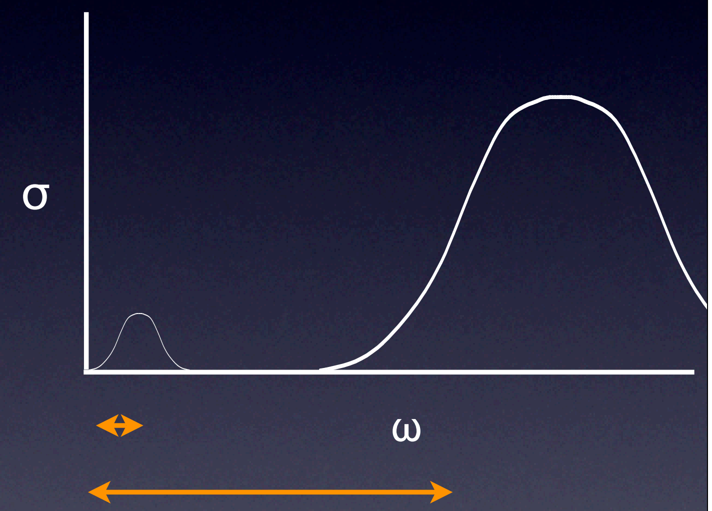
$n_P \sim 1.8 \times 10^{18}/cm^3$

$\xi \sim 13$ nm
$n_P^{-1/3} \sim 9$ nm
$r_{Bohr} \sim 2.5$ nm
$\epsilon \sim 14$ ($\epsilon_{Si} \sim 11.7$)

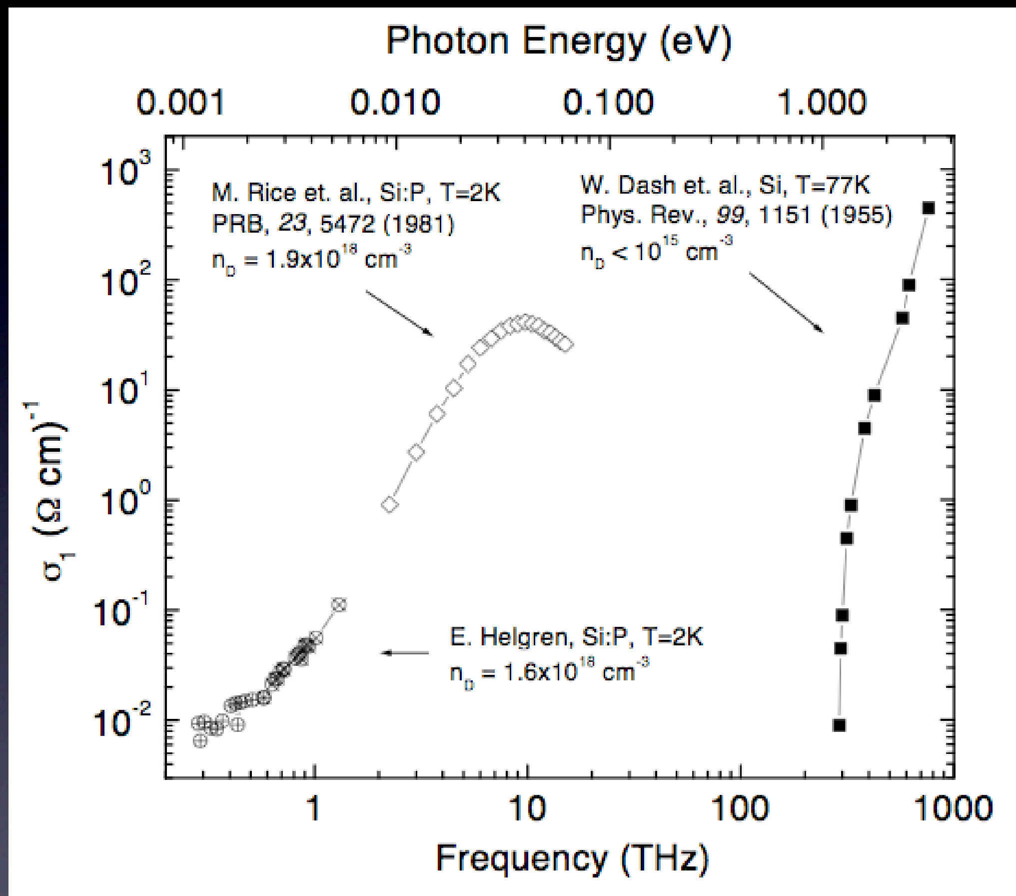
Samples previously used for studies of DC hopping conductivity and AC conductivity in phononless regime

AC Response of Doped Semiconductor





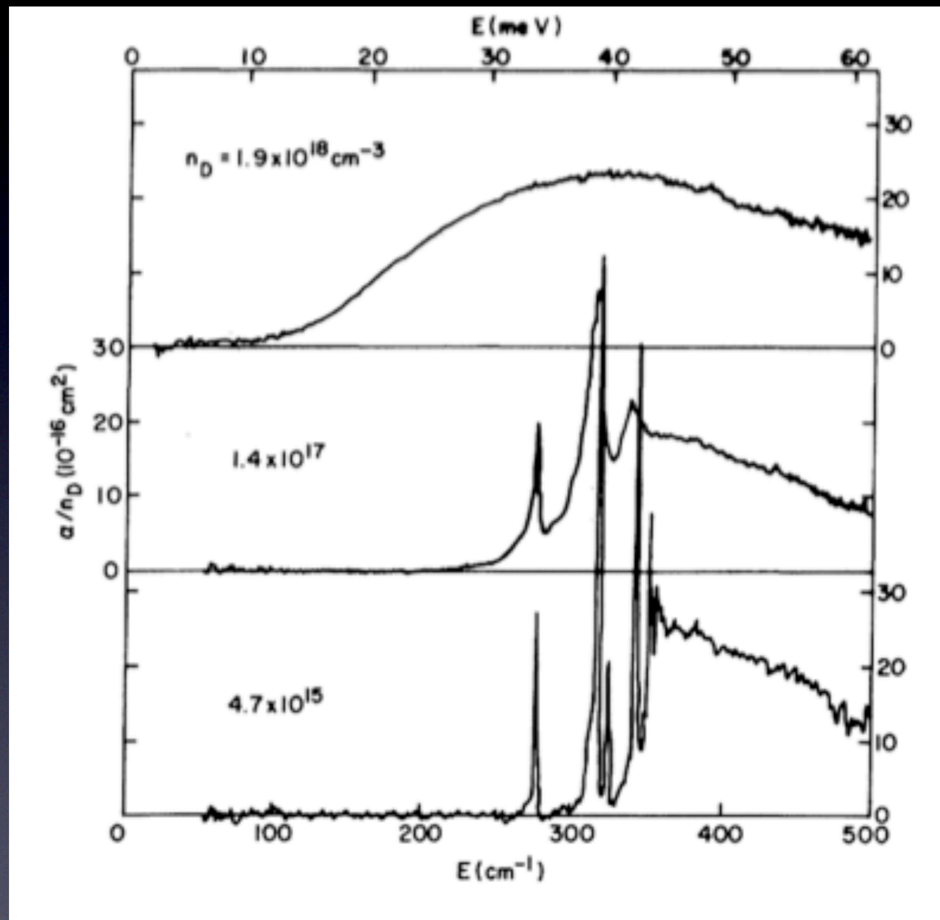
AC response - “Photon assisted”



Helgren, NPA, Gruner PRB 2004

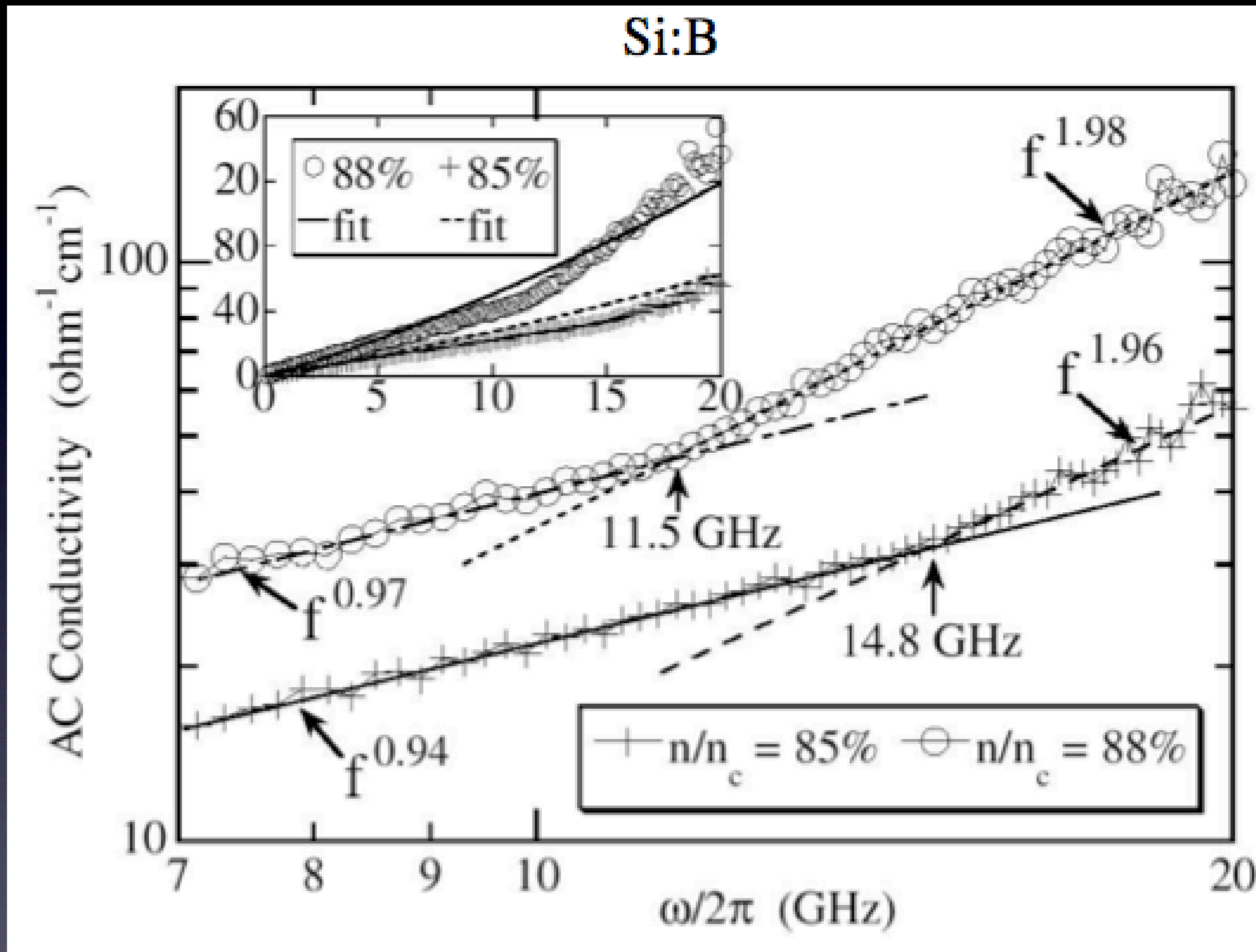
AC response - “Photon assisted”

Doping



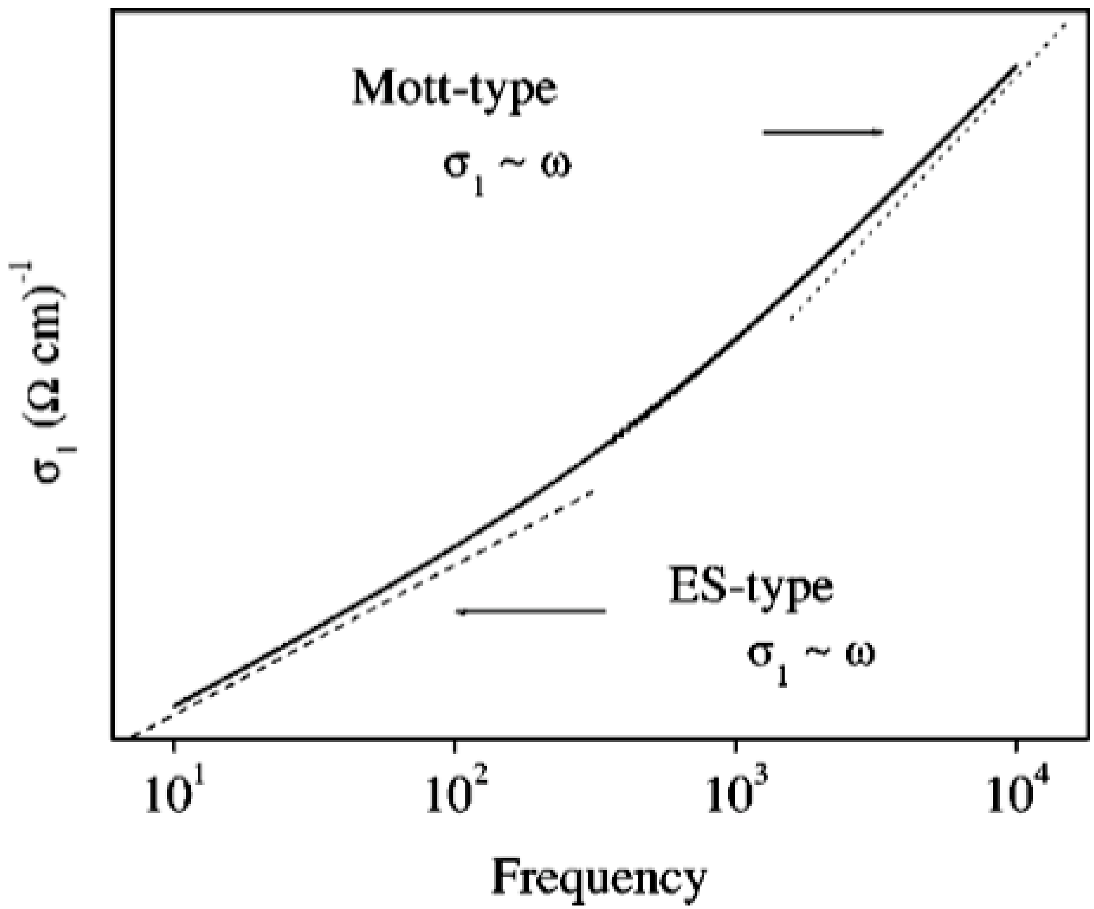
Thomas et al. PRB 1981

AC response - Resonant pair absorption



Lee and Stutzmann PRL 2001

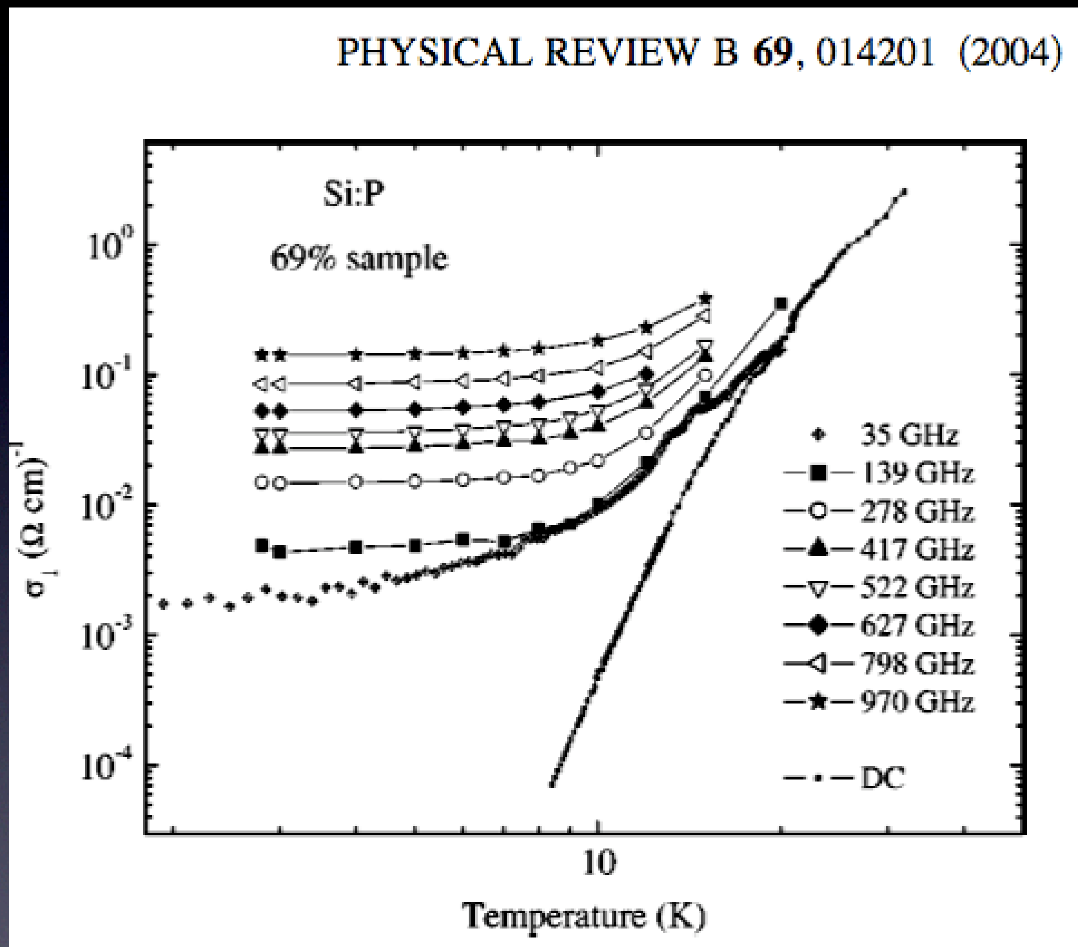
AC response - Resonant pair absorption



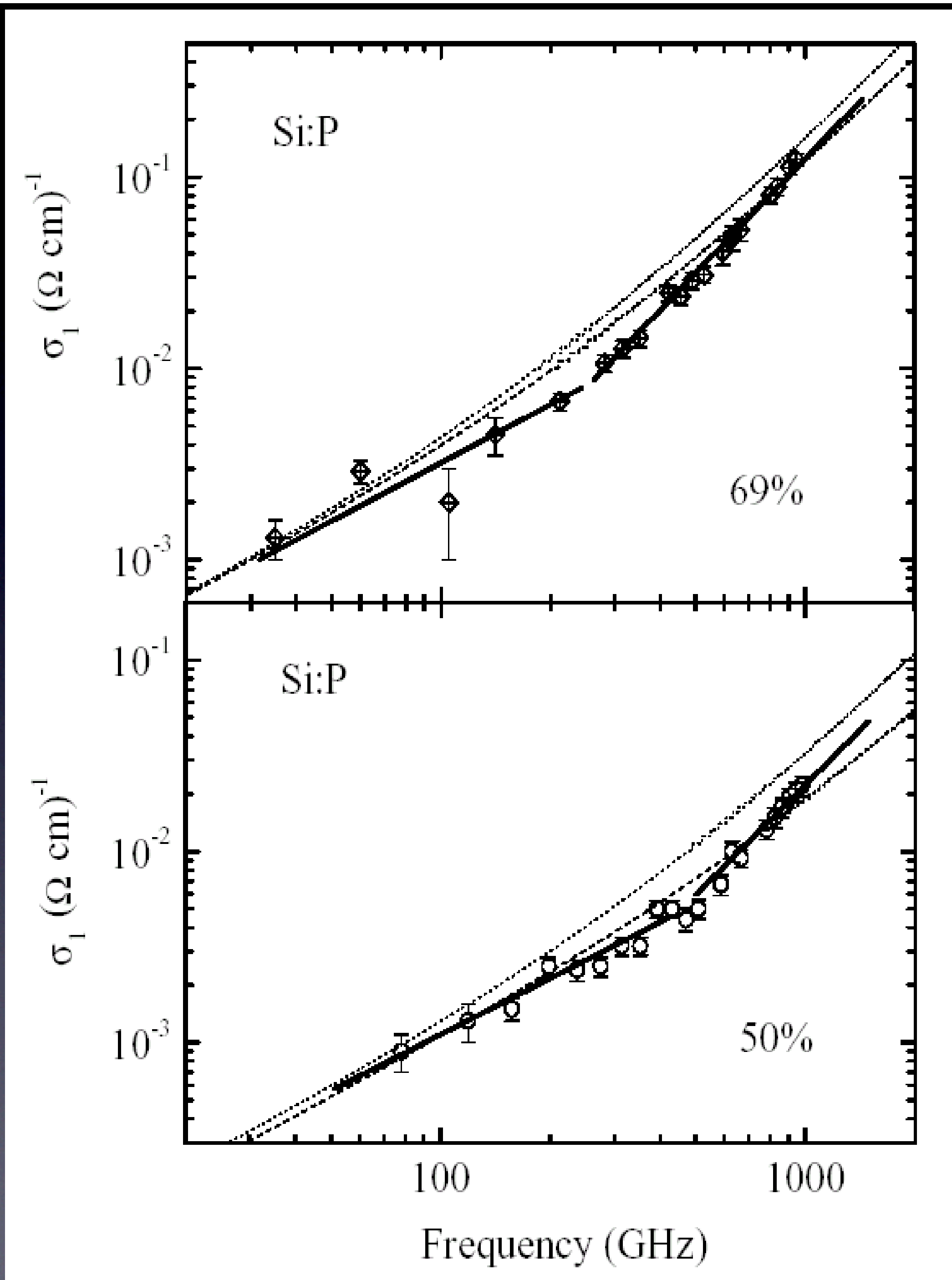
$$\sigma = A\omega\left[\hbar\omega + e^2 \big/ \varepsilon_1 \langle r_\omega \rangle\right]$$

$$\sigma_1 \propto N_0^2 \xi^4$$

Less than ~ 7K is phononless regime



Helgren, NPA, Gruner PRB 2004



AC response

$$\sigma = A\omega\left[\hbar\omega + \frac{e^2}{\varepsilon_1\langle r_\omega\rangle}\right]$$

$\sigma_1 \propto N_0^2\xi^4$

Helgren, NPA, Gruner PRB 2004

PHYSICAL REVIEW B **69**, 014201 (2004)

AC response

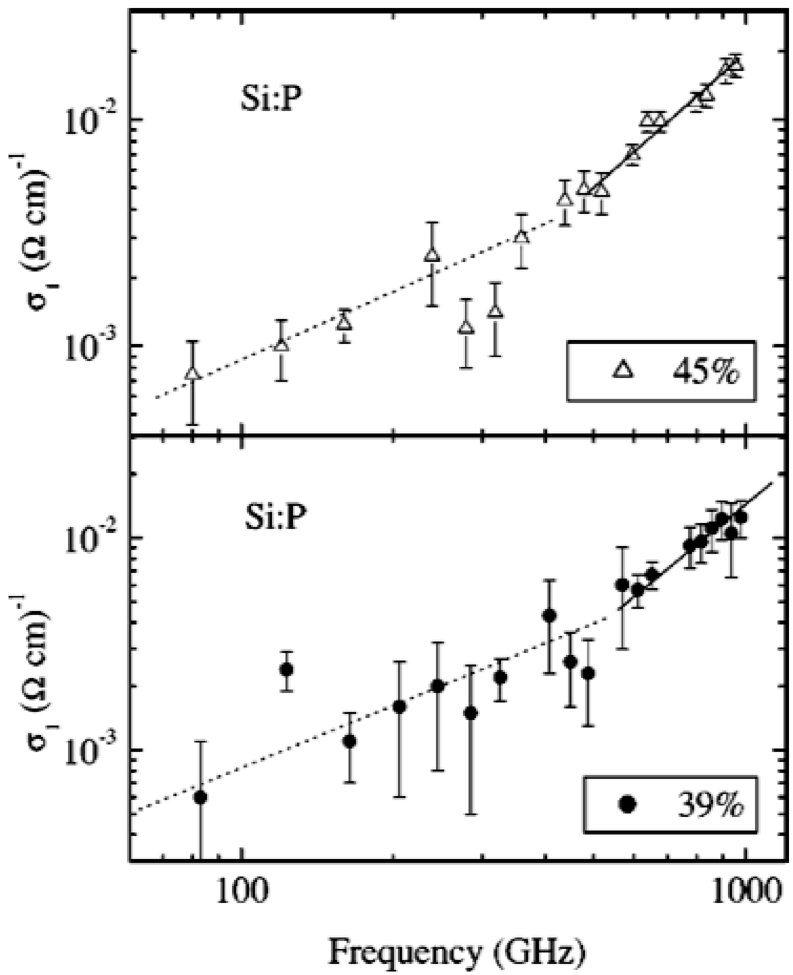
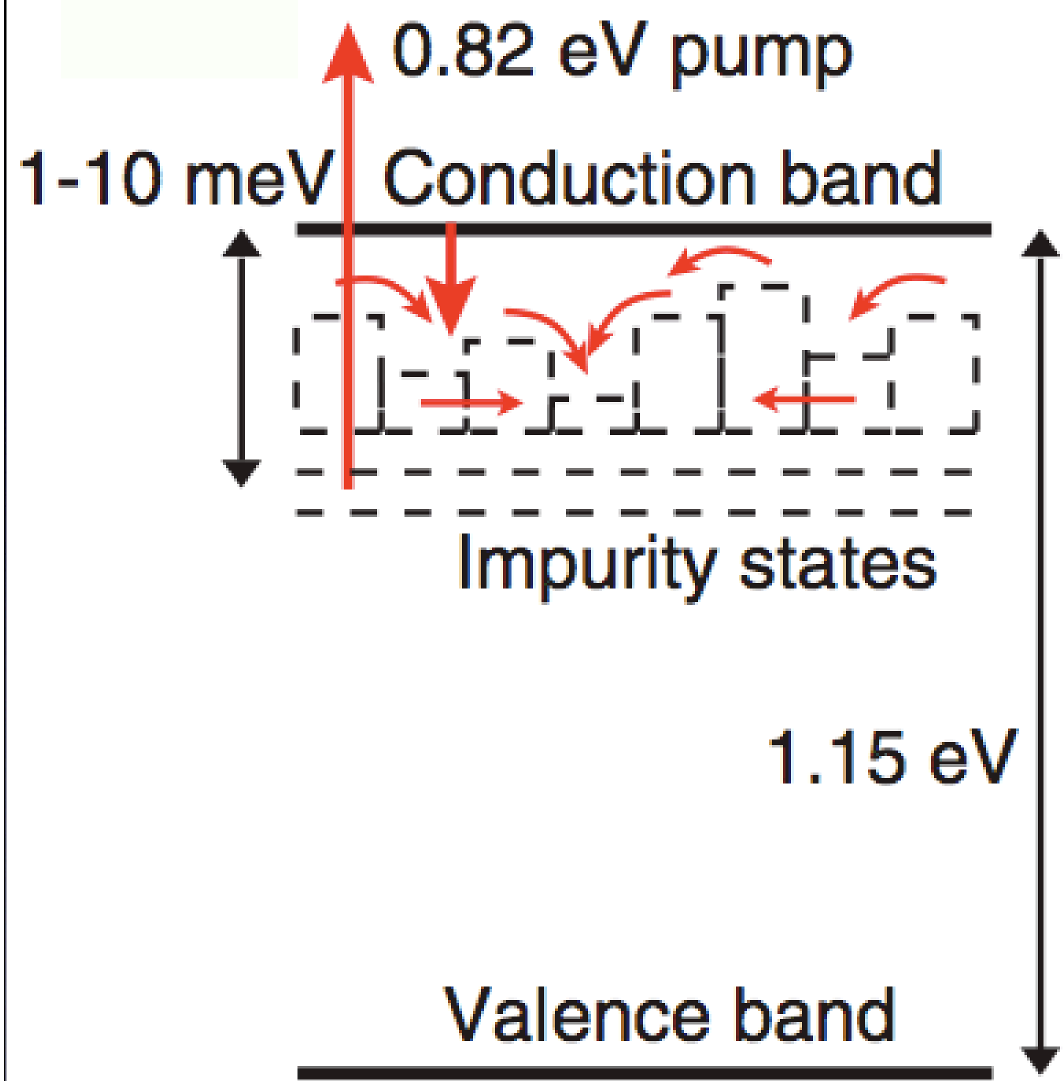


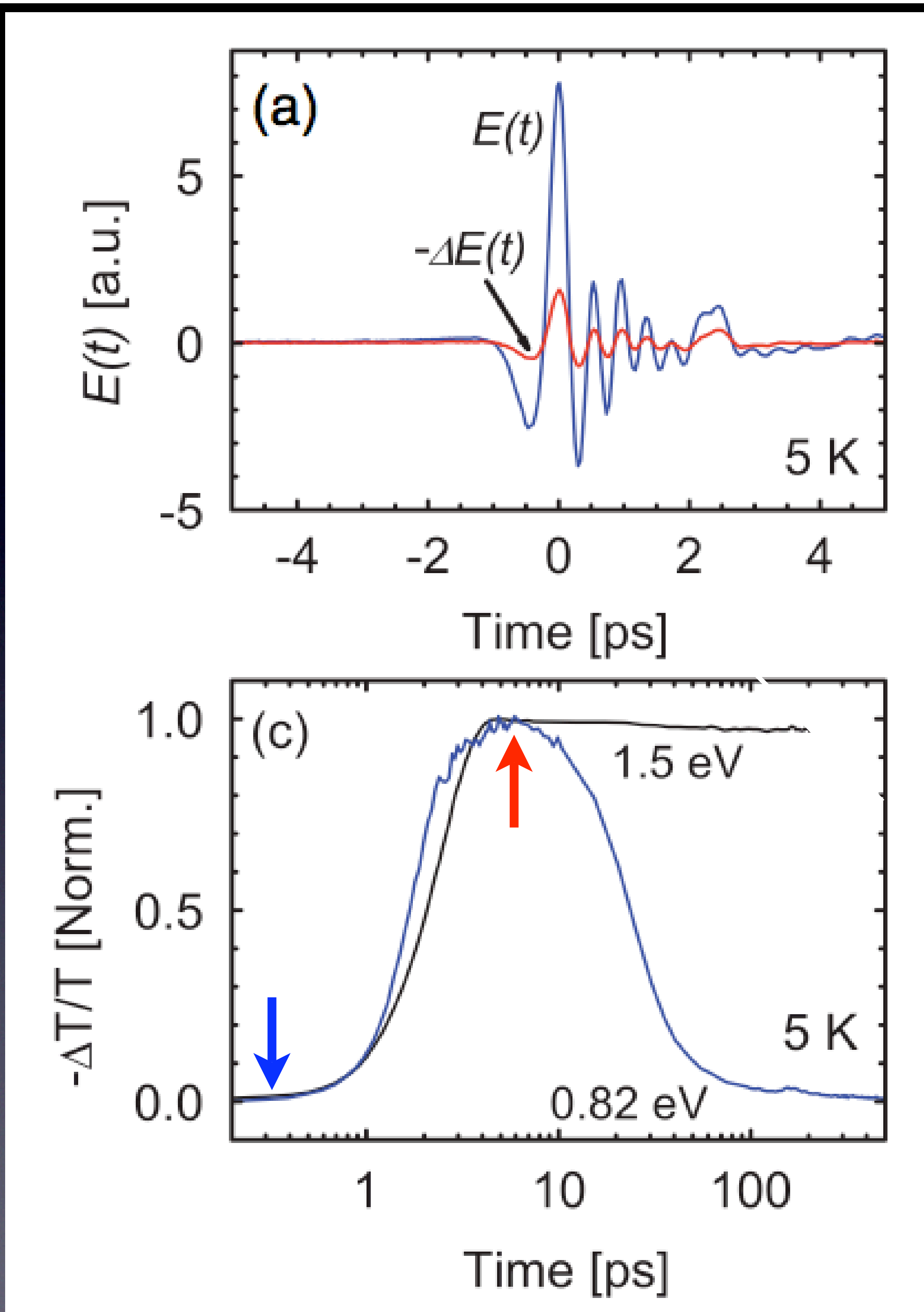
FIG. 7. The measured 2.8 K value of the real part of the complex frequency-dependent conductivity for the Si:P 45% and 39% samples plotted vs frequency on logarithmic axes displaying a crossover in the type of conduction mechanism. The dashed line is a fit to the lower portion of the data and follows a nearly linear power law. The solid line through the upper portion of the data is a fit of those data and follows a nearly quadratic power law.

Helgren, NPA, Gruner PRB 2004

Now PUMP!



Trapping times are generally 3-4 ps

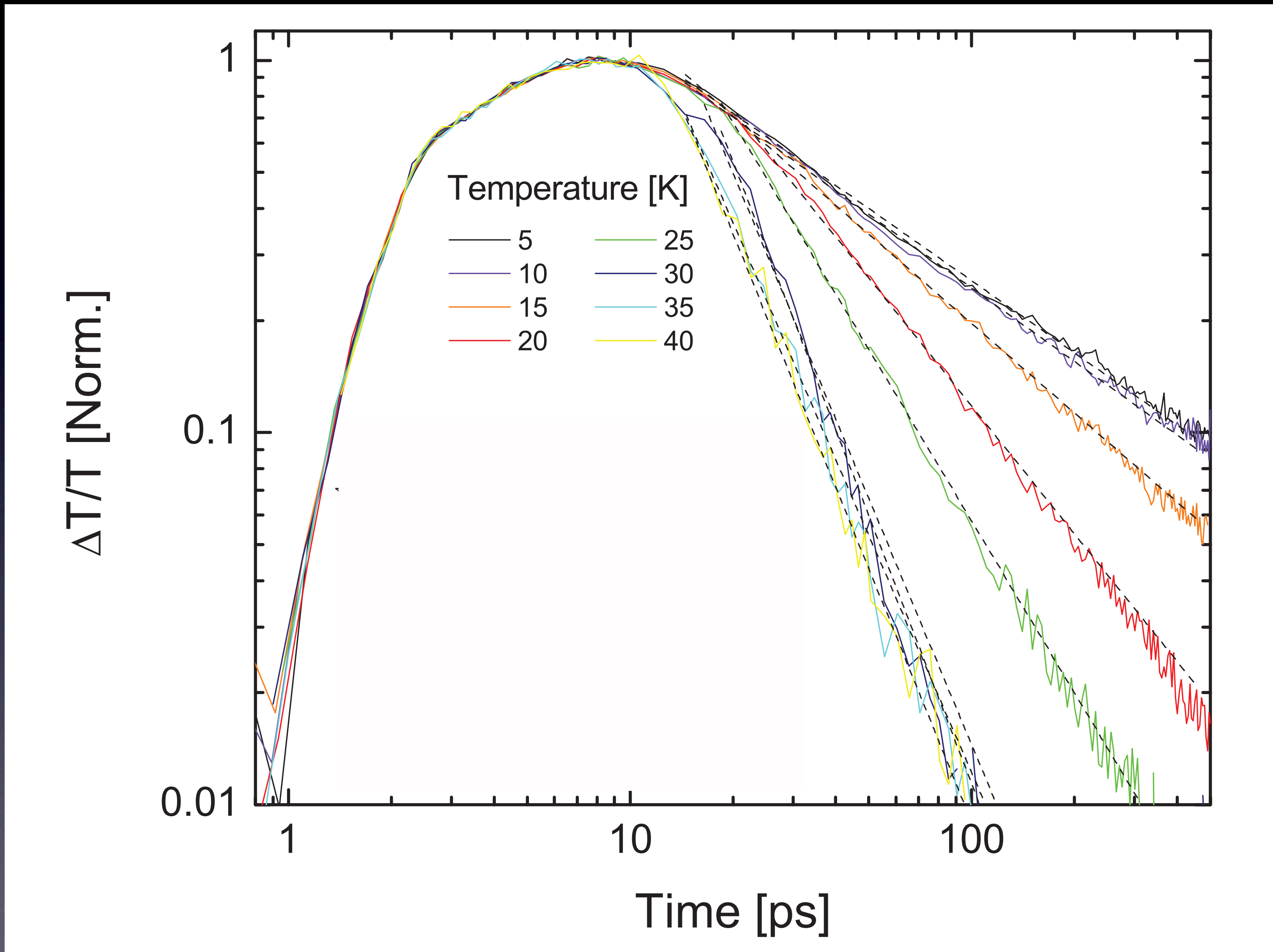


Spectral content of probe pulse peaked at 0.6 THz

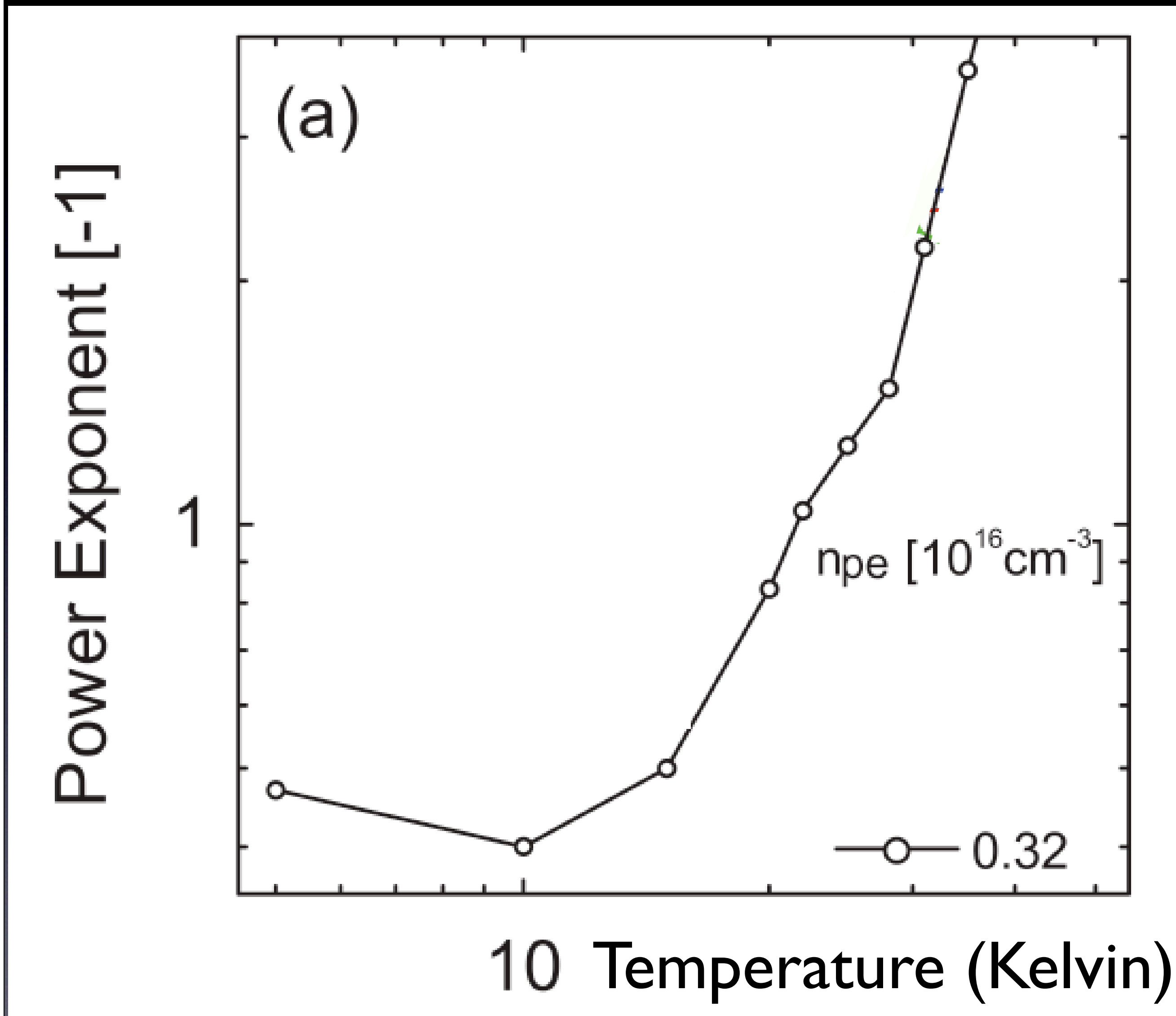
No change in transmission phase; $\sigma \to 0$ @ $\omega \to 0$

Above gap pump is `slow', but not glassy

Transmission vs. pump/probe delay



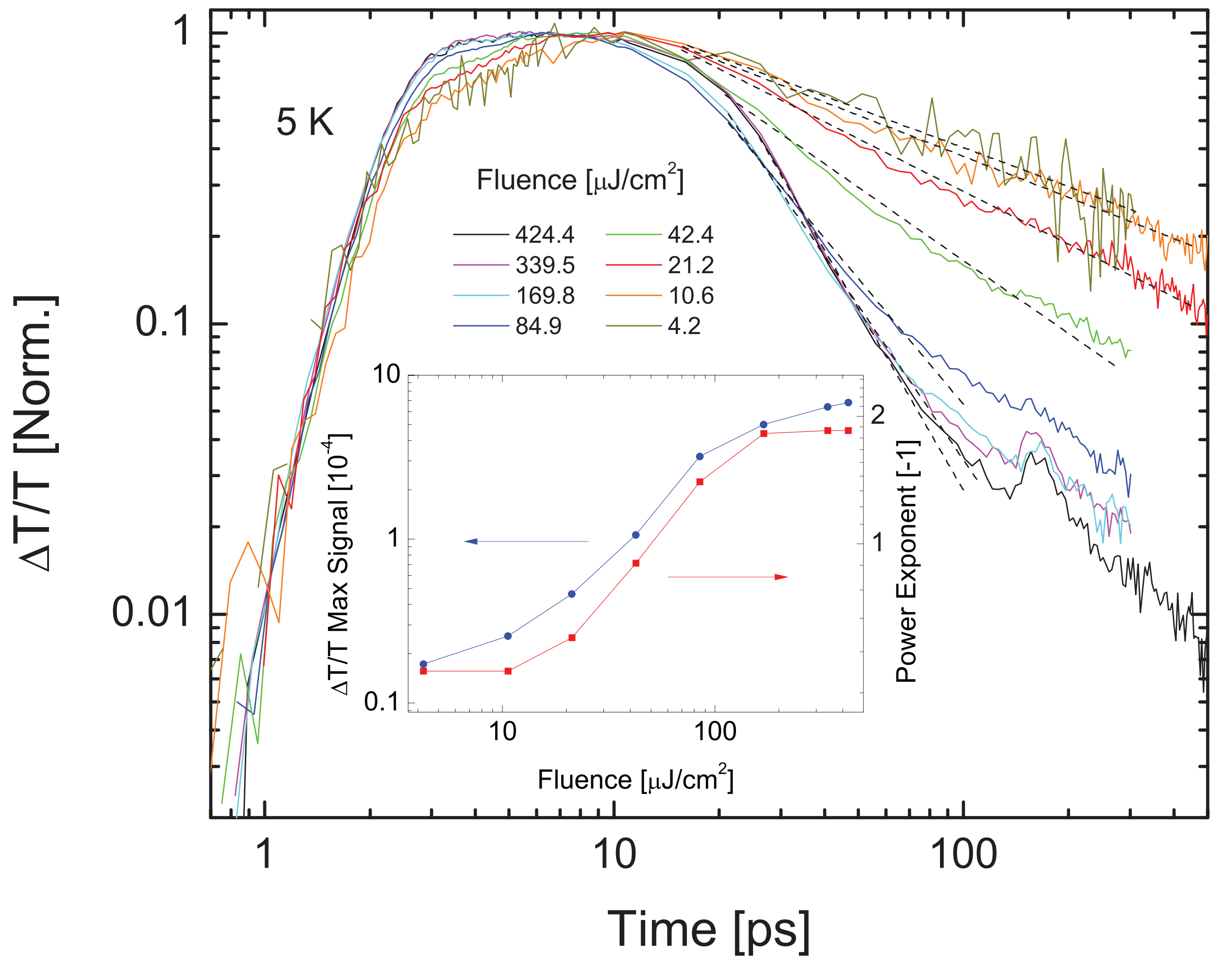
Power law of transmission decay



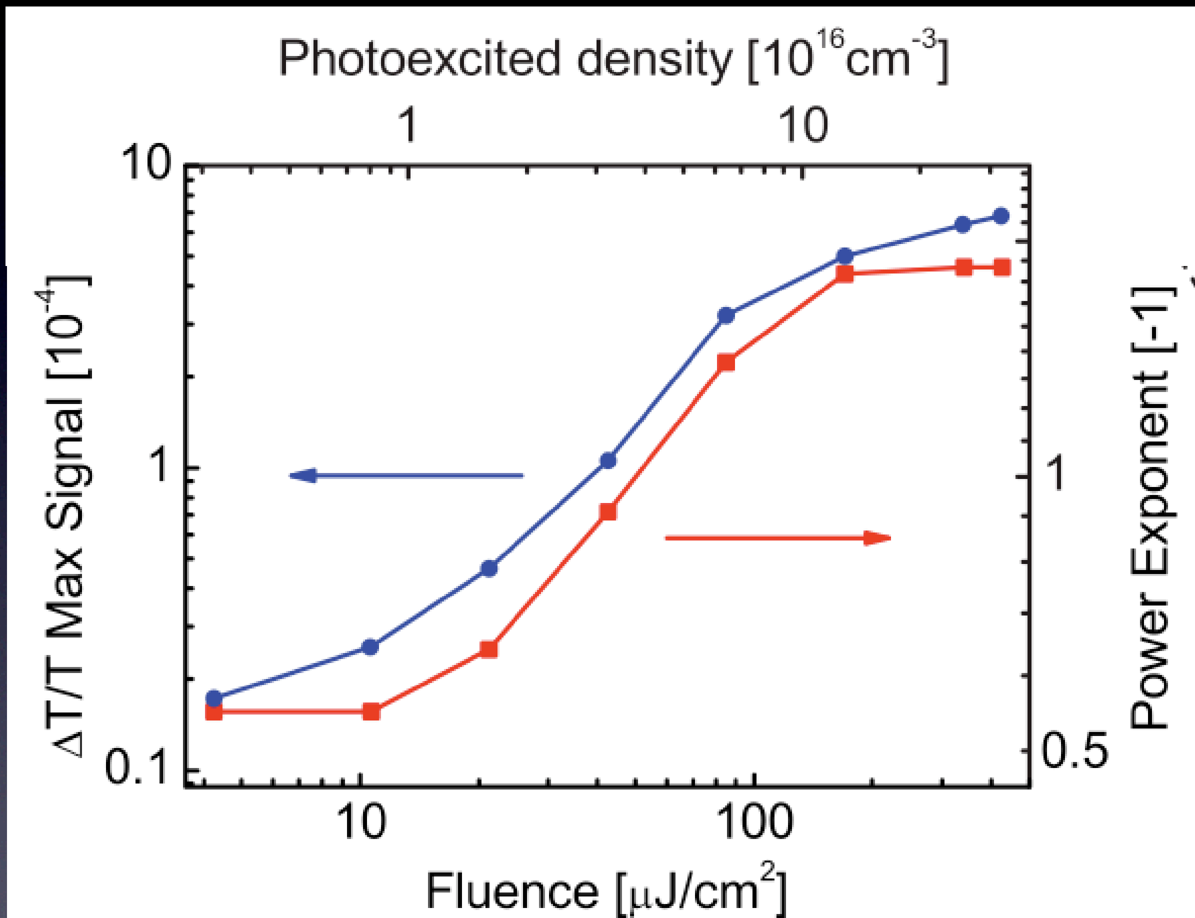
$\sigma(t) \sim T(t) = At^{-\alpha}$

Slow with $|\alpha| < 1$

Transmission vs. pump/probe delay



Power law of transmission decay

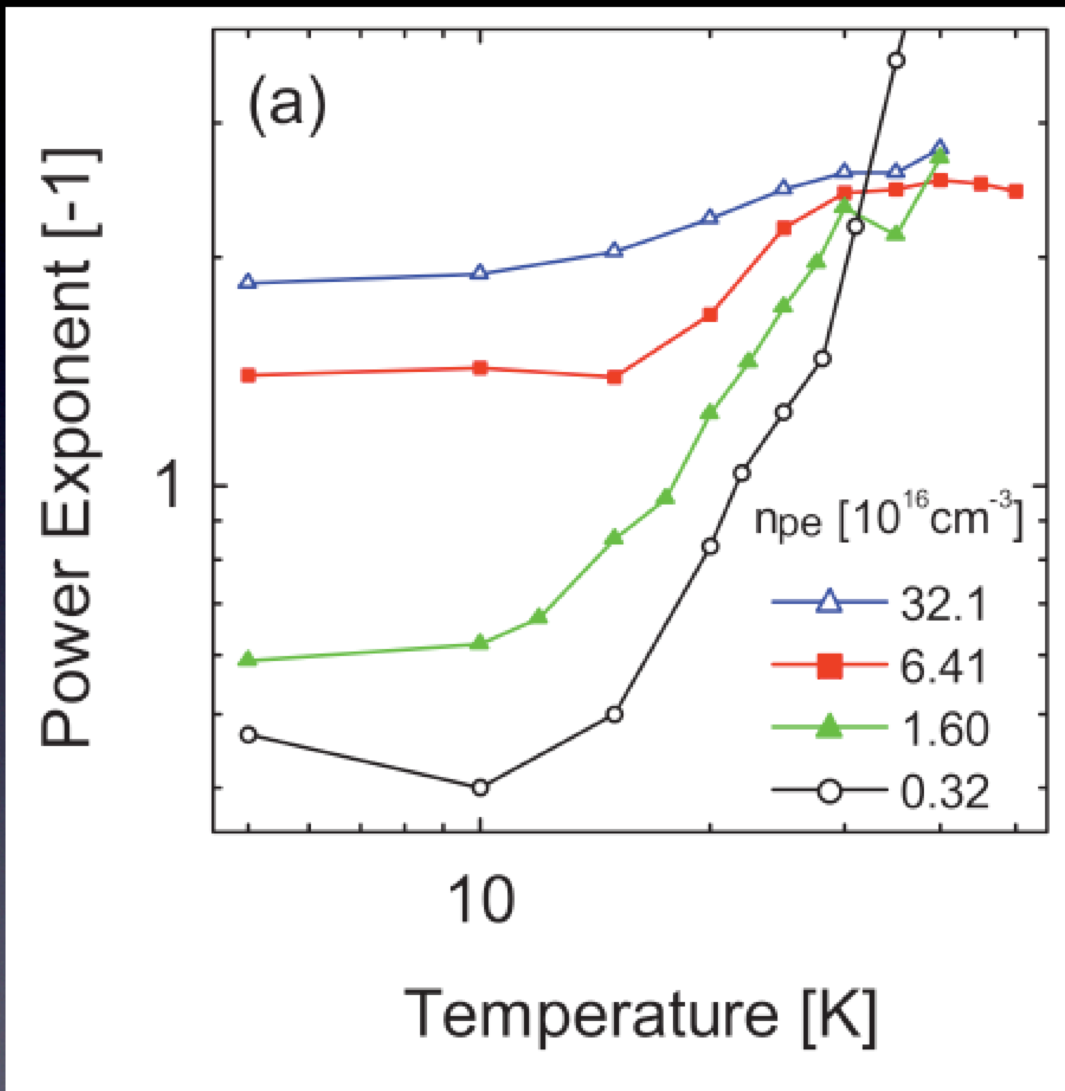


$\sigma(t) \sim T(t) = At^{-\alpha}$

Slow with $|\alpha| < 1$

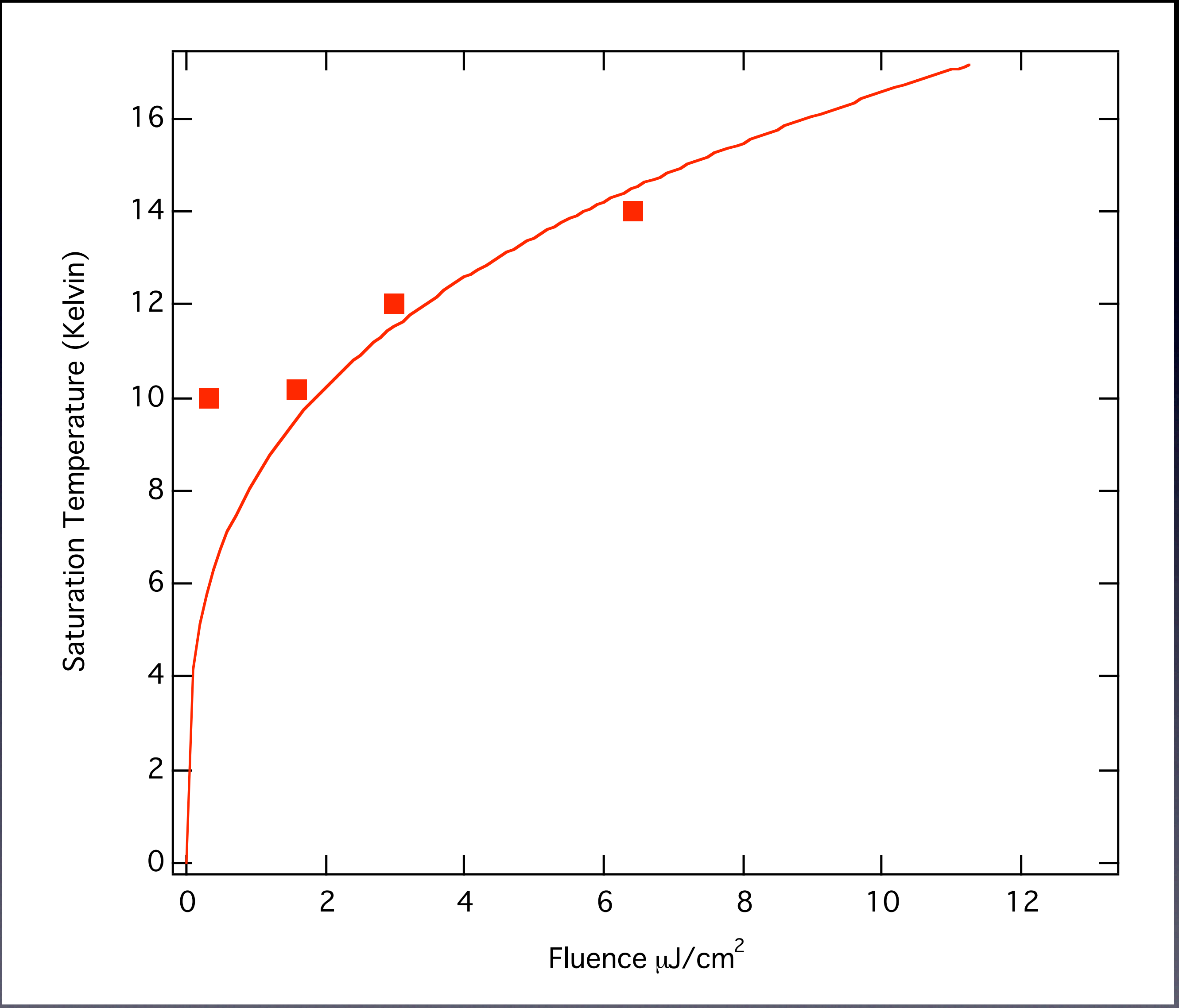
T = 5 K

Power law of transmission decay



$\sigma(t) \sim T(t) = At^{-\alpha}$

Quantum relaxation regime?



@ Low T and fluence ΔT/T falls to 30% in 200 ps

$$\tau_{phonon} = \frac{1}{\omega_D} e^{\left(\frac{T_{ES}}{T}\right)^{1/2}} \approx 9 \text{ picosecond}$$

phonon assisted

$$\tau_M = \rho_{DC}/4\pi\epsilon_1\epsilon_0 \approx 0.9 \text{ millisecond}$$

Maxwell time

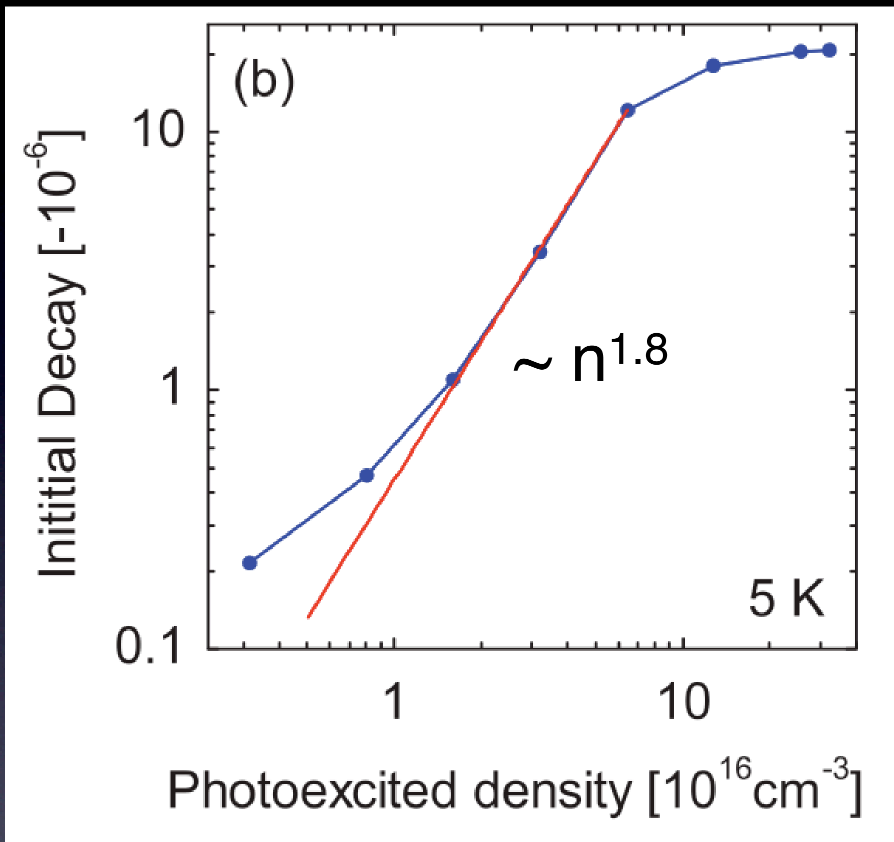
$$\tau_{M,AC} = \rho_{AC}/4\pi\epsilon_1\epsilon_0 \approx 14 \text{ picosecond}$$

Maxwell time @ 600 GHz

Relaxation time much longer than naive scales

Totally unrelated to Maxwell time

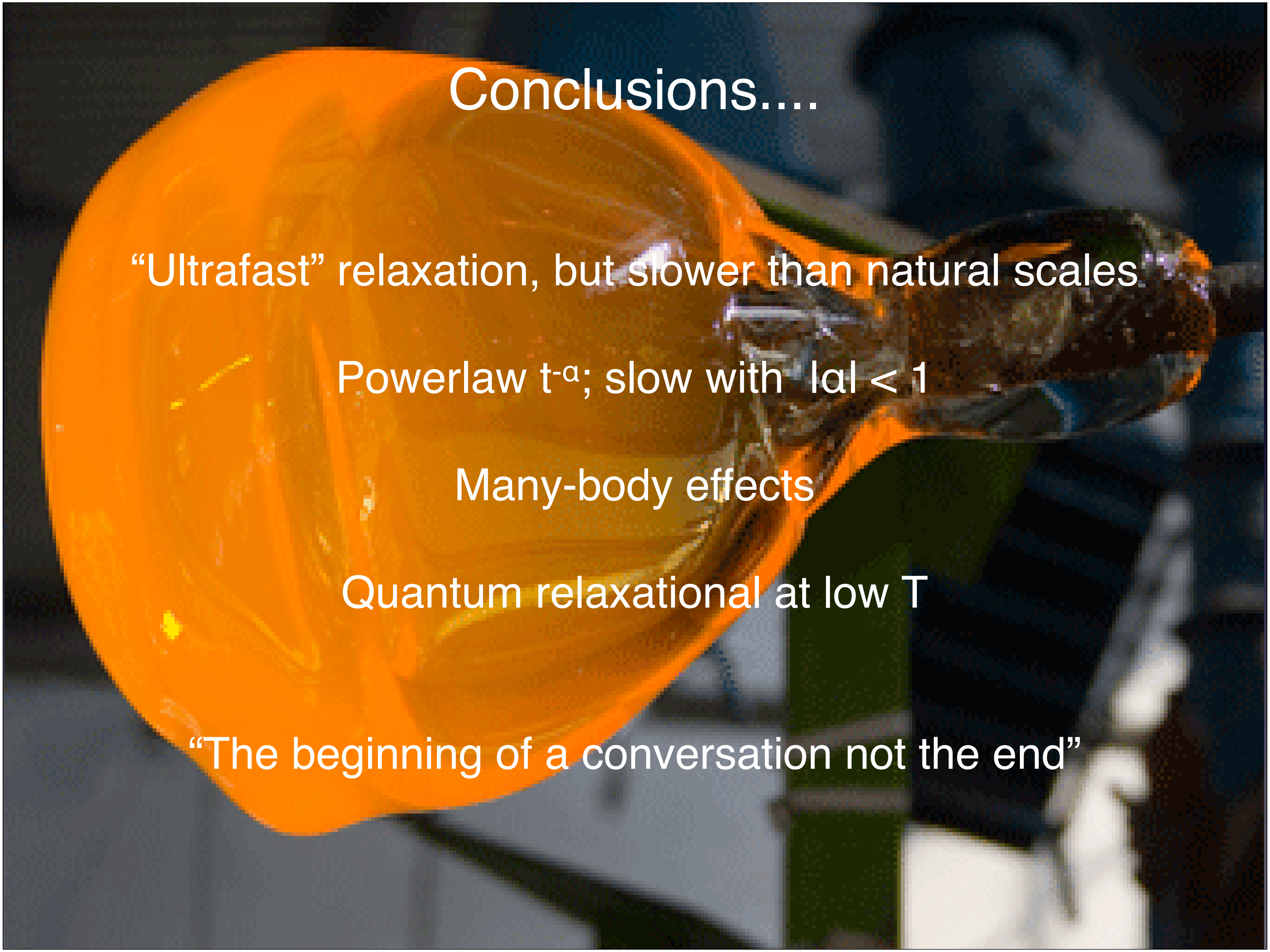
Multi-particle relaxation



Rate goes as power > 1;
Evidence for multi-particle relaxation processes

$$\dot{n} = I_{qp} + 2N\gamma_{pc} - \beta n^2$$

$$\dot{N} = I_{ph} + \beta n^2/2 - \gamma_{pc}N - (N - N_{eq})\gamma_{esc}$$


Conclusions....
“Ultrafast” relaxation, but slower than natural scales
Powerlaw $t^{-\alpha}$; slow with $|\alpha| < 1$
Many-body effects
Quantum relaxational at low T
“The beginning of a conversation not the end”