Time-resolved far-IR studies

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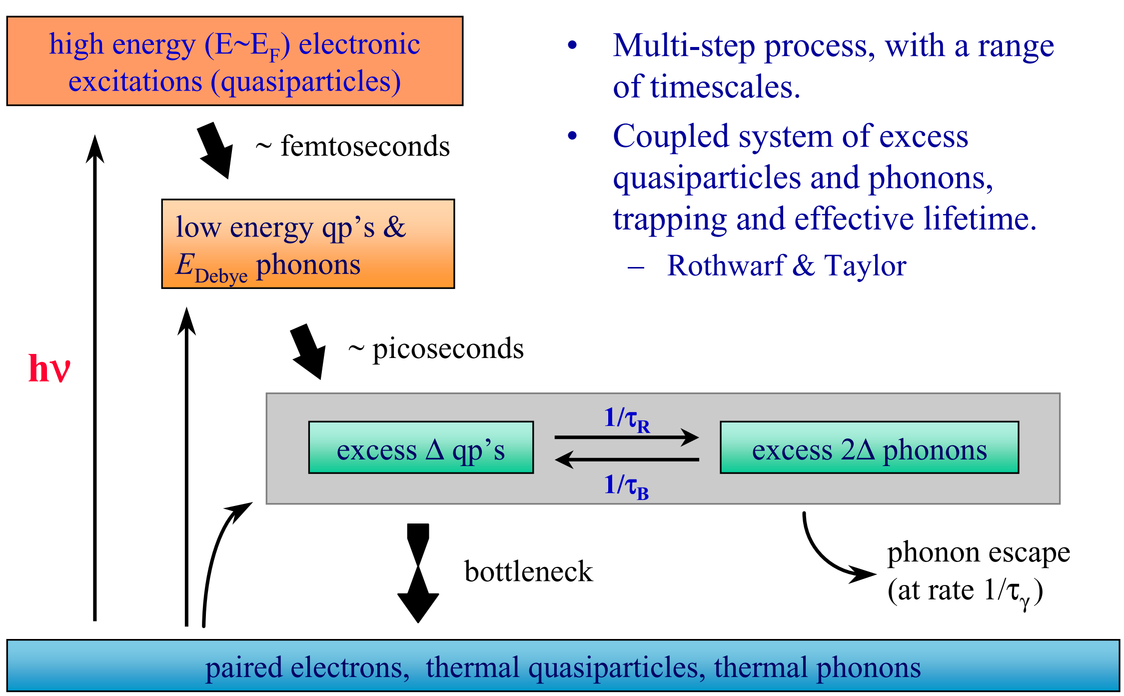
Funded by DOE contracts DE-FG02-02ER45984 at UF, and DE-AC02-98CH10886 at BNL

OUTLINE

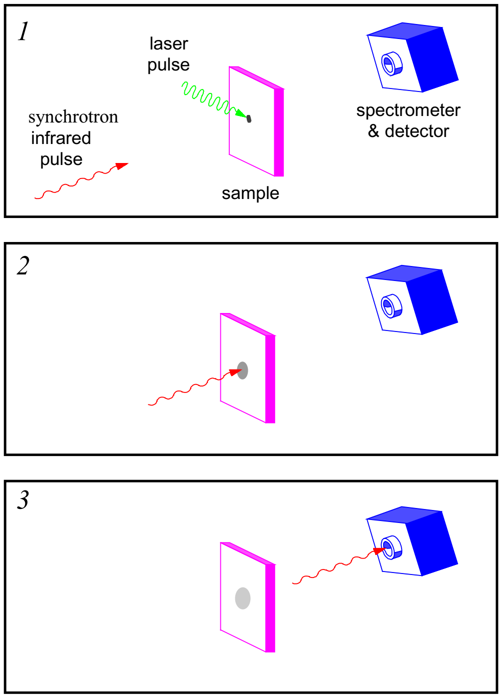
- Non-equilibrium state and relaxation process in superconductors
- Pump-probe technique
- MoGe
- Time-dependent relaxation of quasiparticles
- Conclusions

Relaxation processes in superconductors



- Multi-step process, with a range of timescales.
- Coupled system of excess quasiparticles and phonons, trapping and effective lifetime.
  - Rothwarf & Taylor

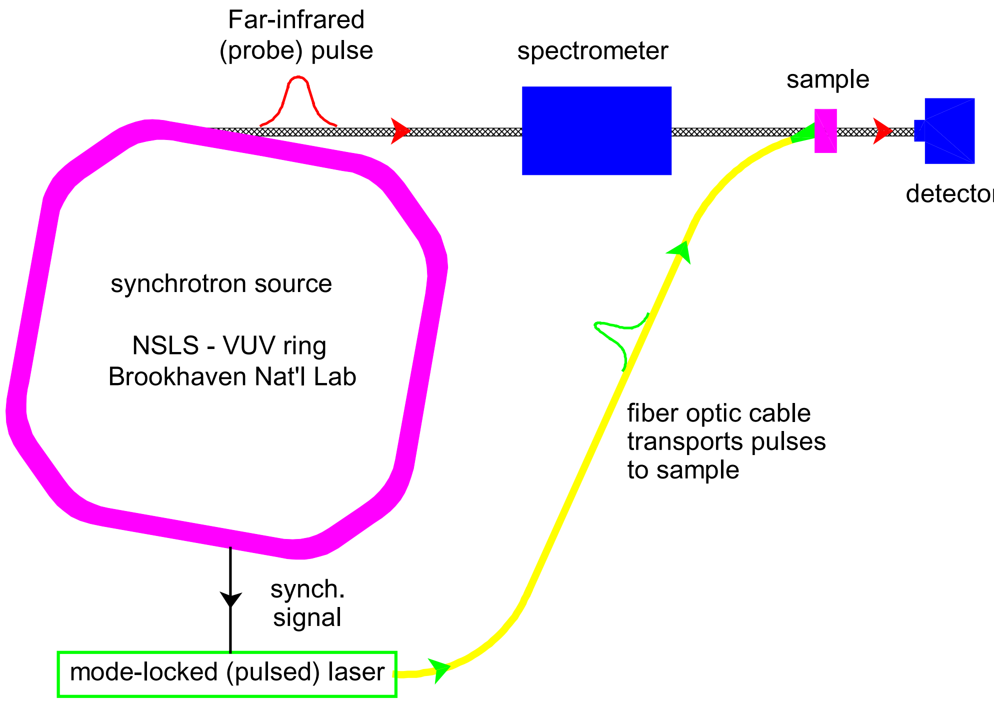
Pump–probe method for time–resolved FIR



1. Laser pulse creates photoexcitations in sample, which subsequently evolve with time.
2. After time Δt, broadband (continuum) IR pulse arrives and is partially absorbed (or reflected) by excitations.
3. IR pulse analyzed with a spectrometer, extracting details of excitations at a time Δt after their creation.

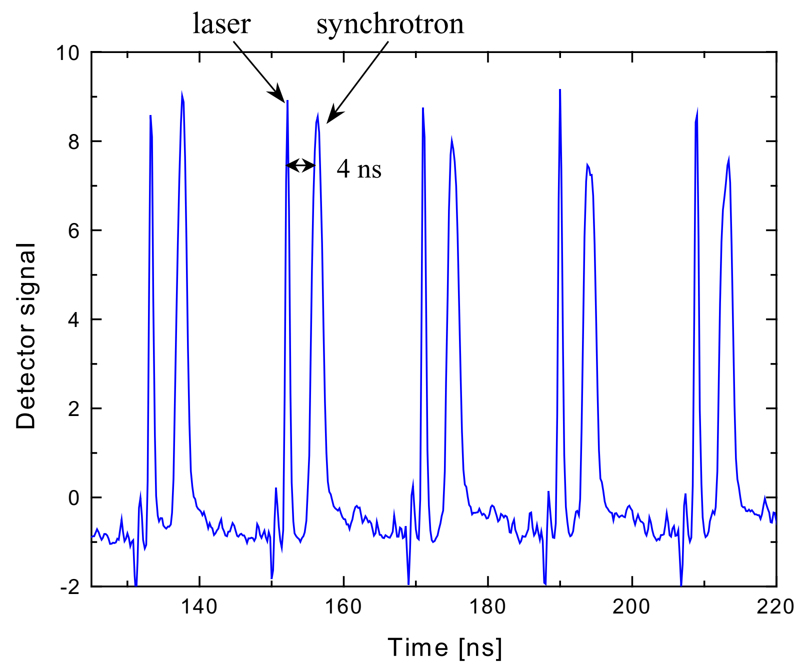
- Cycle repeats at high (MHz) repetition rate.
- Photoexcitation evolution determined by measuring at a variety of Δt's.
- Employs "conventional" spectroscopy using high-sensitivity (slow-response) detectors.

Pump-probe with laser & synchrotron pulses



Pulses at specimen's location

- Synchronized laser & storage ring pulses.
- Measured in sample chamber of U10A spectrometer.
- with Ge APD (near IR detector, ~1 ns response). Useful for locating "zero" delay point.
- Here, the delay is 4 ns.



α-MoGe thin films

FILMS: Grown by RF sputtering
Ge buffer layer
Thickness measured with quartz thickness monitor.

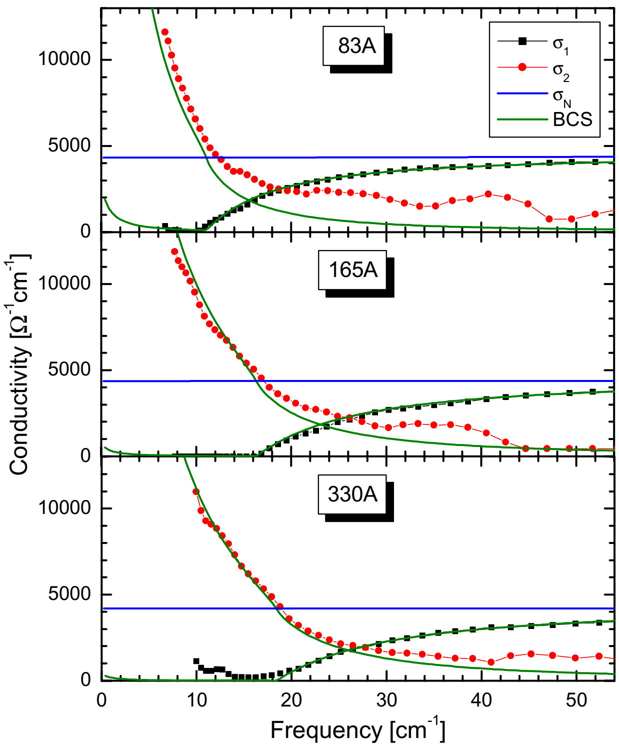
SUBSTRATE: sapphire (r-cut)
1 mm thick
$n_{subs} \sim 3.05$

KNOWN: T_c bulk is about 7.2 K.
T_c varies linearly with thickness and $1/R_\square$.
mean free path is of order 1-3 Angstroms

- A strong reduction of T_c with increasing $R_\square$ is attributed to localization and related changes in the Coulomb interaction.
- α-MoGe serves as a model system for studying the interplay between superconductivity and disorder.
  - $\rightarrow R_\square$ is the relevant measure of disorder in 2D.

J.M. Graybeal and M.R. Beasley, *Phys. Rev. B* **29**, 4167(1984)

Conductivity of MoGe Films at 2.2K



Conductivity of a thin film on a thick substrate from transmittance and reflectance:

- Algorithm based on the approaches of Palmer and Tinkham and also Glover and Tinkham

R.E. Glover, III and M. Tinkham, *Phys. Rev.* 108, 243(1957)

L.H. Palmer and M. Tinkham, *Phys. Rev.* 165, 588(1968)

- σ_1 fits well to BCS for all three MoGe films.
- σ_2 has correct lineshape, ~$1/\omega$, but is above BCS, especially at higher frequencies.

Now define:

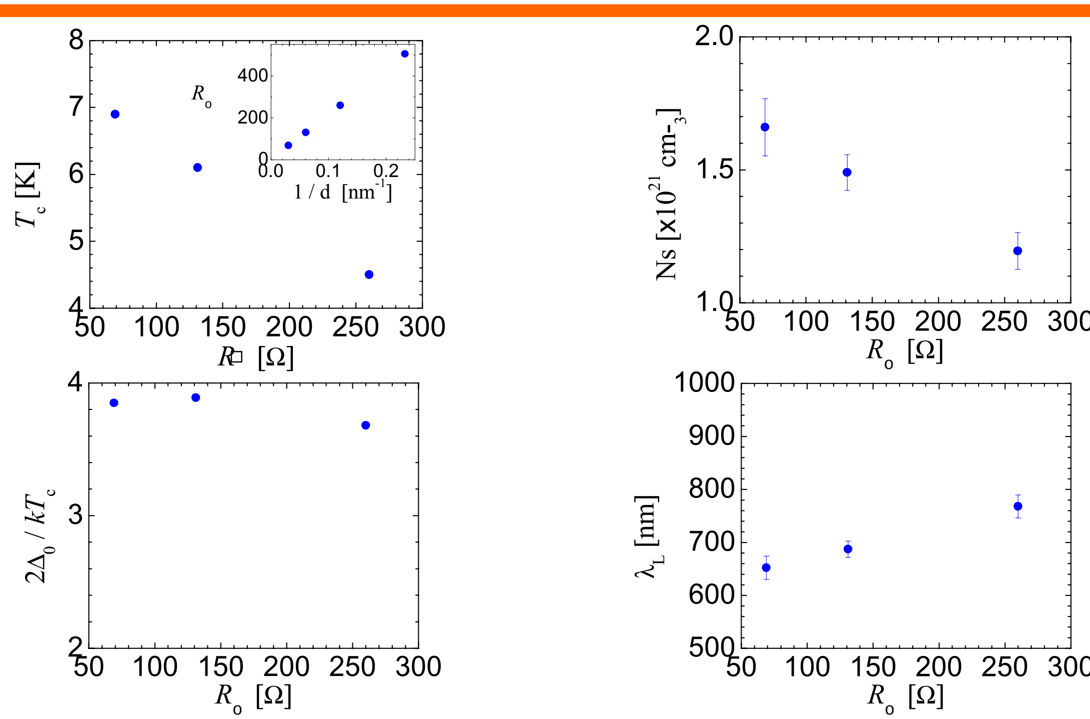
Superfluid density:

$$\boxed{N_s(m/m^*) = mV_c\omega\sigma_2 / e^2}$$

London penetration depth:

$$\boxed{\lambda_L = c / [4\pi\omega\sigma_2]^{1/2}}$$

How parameters depend on R_o

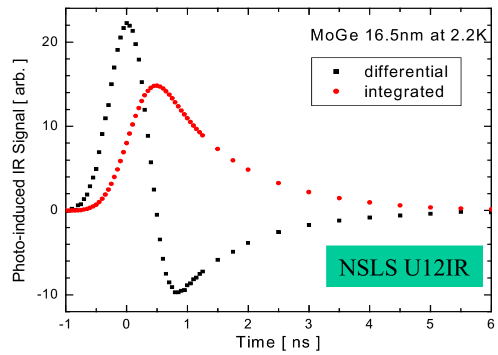


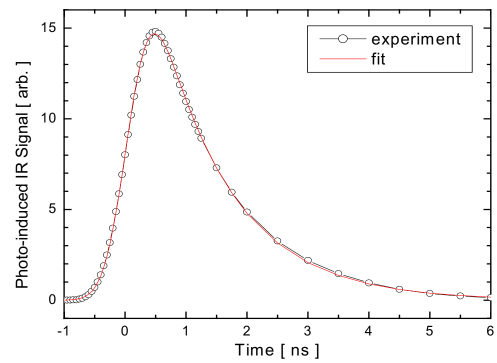
- T_c varies strongly with R_o and the superfluid density.
- $2\Delta_0 / kT_c$ does not depend strongly on R_o.

Time–dependent relaxation measurements

- Differential Technique:
  - pump-probe delay “dithered”.
  - differential transmittance signal (spectral average) for a range of delay time.
  - time-dependent relaxation of excess quasiparticles by integration.
- Relaxation Behavior:
  - convolution of simple exponential decay and Gaussian synchrotron pulse.
  - decay time ~ 1 ns.
  - time-resolution determined by synchrotron pulse width (> 300 ps).

$$\Delta T = \frac{1}{2} A \exp\left(\frac{w^2}{4\tau^2} - \frac{t-t_0}{\tau}\right)\left(1 + erf\left(-\frac{w}{\tau} + \frac{t-t_0}{w}\right)\right)$$





Temperature dependence of relaxation time

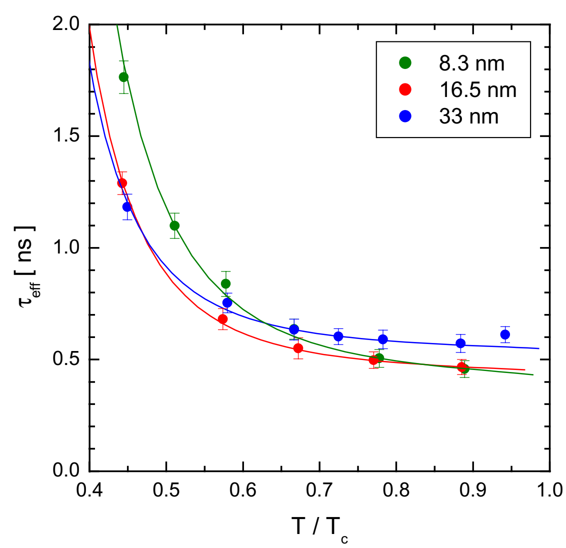
$$\tau_R(0) = (1+\lambda)\hbar / 2\pi b\left(kT_c\right)^3$$

$$\tau_B(0) = \hbar N_\Omega / 4\pi^2 N(0)\left\langle \alpha^2 \right\rangle_{av} \Delta_0$$

$$\tau_{eff} \approx \tau_\gamma + (1/2)\tau_R(1 + \tau_\gamma / \tau_B)$$

- Relaxation times for MoGe films:

d [nm]	τ_γ [ps]	$\tau_R(0)$ [ps]	$\tau_B(0)$ [ps]	$\tau_R(0)/\tau_B(0)$
8.3	420	370	105	3.50
16.5	450	150	79	1.87
33	550	100	75	1.35



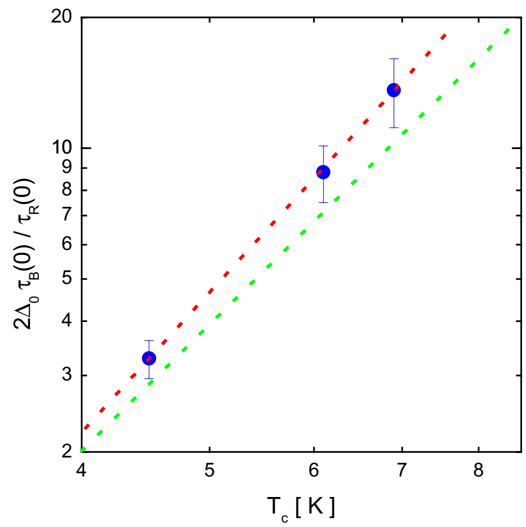
S. B. Kaplan et al.

Pair–breaking and recombination ratio

- Prediction by the theory (assuming that material parameters are the same for all three films):

$$\frac{2\Delta_0 \tau_B(0)}{\tau_R(0)} \propto T_c^3$$

- Experiment:
  - The ratio ~ T_c^3.
  - Support the analysis of Kaplan et al.
  - Possible deviation due to invalid assumptions of the theory (e.g. weak-coupling, constant α^2, simple Ω^2 dependence of phonon density of states, etc).



Conclusions

- Used pump-probe technique to follow relaxation of non-equilibrium superconducting a-MoGe films
- All three films fit dirty-limit BCS
- Our results are generally consistent with the theory by Kaplan et al.
- The effect of reduced thickness in these samples is to depress T_c and the superfluid density.
- At the same time, the normal-state conductivity and mean free path appear unchanged
- Indeed, it seems that, other than T_c, none of material parameters are changing with thickness.

Needs of a new facility

- Faster FIR pulses – 1-10 ps
- Excellent FIR S/N ~10^5/rHr (by power or stability or detector)
- Spectral coverage 1-100,000 cm^{-1}
- Modest resolution (0.5 cm^{-1} at 20 cm^{-1} is plenty)
- Low T capability: 1.7-300 K
- Range of pump wavelengths (FIR, MIR, NIR, VIS, UV)
- Range of pump fluences (up to ~100 mW)
- Magnet?

Marshall Onellion and Prasenjit Guptasarma, Wisconsin

(1) **Ferromagnetic Semiconductors**
- GaN:Mn, (ferromagnetic, Curie temperature > 300K)
- Pump-probe with a pump slightly larger than the GaN:Mn bandgap. (Need ~ 3 to ~ 5 eV)

(2) **Quantum confined Laser media**
- III-V compounds.
- Carrier relaxation is critical to the lasing medium (determines population inversion)
- Pump at the bandgap, 800- 950 nm (Ti:sapphire) or 3-5 m (OPA).
- Use THz as a probe of the carrier relaxation.

(3) **Dual phase transitions: Multiferroics- Ferroelectric and Ferromagnetic**
- Exhibit coexistence of two phase transitions: ferroelectricity + ether antiferromagnetism or ferromagnetism.
- Example material: $BiFe0_3$:Tb
- Pump carriers across the bandgap, using variable pump light polarization, and
- Study carrier thermalization and relaxation, in both the ordered and disordered states
- Need pump laser in the photon energy range of 3-5 eV.

(4) **Cuprate superconductors**
- Measure versus temperature and applied magnetic field,
- Measure transient change from photoexcitation.

(5) **Ruthenates: Sr_2RuO_4 and homologous series**
- Sr_2RuO_4: T_c of 3 K (special case) or lower; 1.5 K more typical. Need a ^{3}He cooling system.
- Normal state differs qualitatively between Sr_2RuO_4 and $SrRuO_3$
- Sr_2RuO_4 has Fermi Liquid behavior over 1.5-30 K, crossover to non-Fermi liquid characteristics above ~ 25-30K.
- Measure THz absorption versus temperature

(6) **Magnetic nanowire arrays**
- Make magnetic nanowire arrays (Ni, Fe, Co, alloys) inside of alumina honeycomb arrays
- Wire diameter down to ~ 5 nm to ~ 400 nm; 10-15% of the volume is wires
- Measure the THz absorption as a function of temperature and applied magnetic field
- Transient demagnetization studied by pump/probe (Ti:sapphire)

James Lawler, Wisconsin

- Lighting consumes approximately 25% of all electrical power.
- Discharge sources typically 5 to 10x more efficient than incandescent sources
- High Intensity Discharge (HID) lamps, High Pressure Sodium (HPS) and Metal Halide (MH-HID), are in widespread use today.
- HID lamps operate in local thermodynamic equilibrium
- Total pressures: 1 to over 200 bar,
- Electron densities from 10^{15} - 10^{18} cm^{-3} (Plasma frequencies 0,3-10 THz)
- Diagnostic experiments on HID lamps are extremely challenging.
- Synchrotron radiation has the needed spectral radiance to "outshine" these very intense plasmas across broad spectral regions. (Optical and UV absorption at Stoughton / K-shell X-Ray at APS
- Intense Terahertz radiation would be extremely valuable both for electron density measurements and for measuring the opacity of these lamps due to electron-atom inverse bremsstrahlung.
- Lamps are compact and well suited to experiments at remote sites.