Excitation Intensity Dependence of Ultrafast Carrier Dynamics in GaAs

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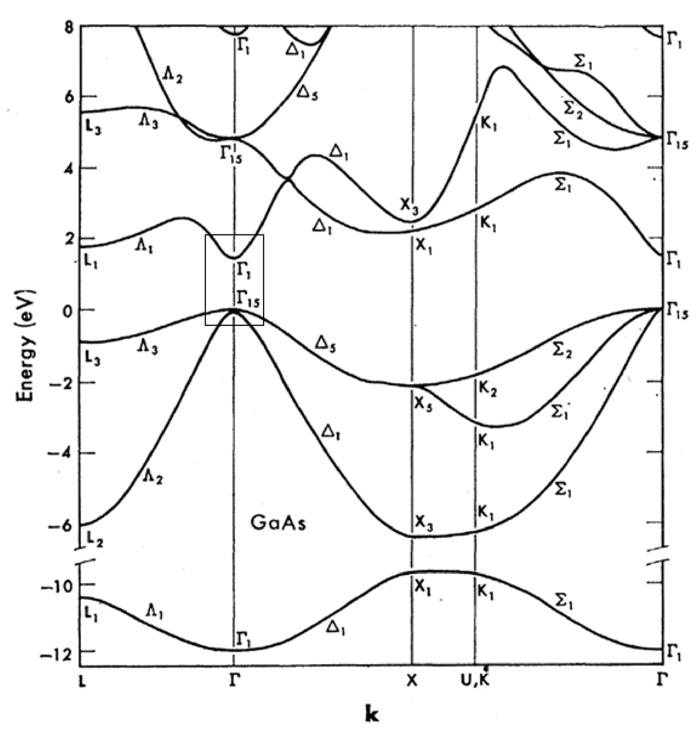
Introduction :

While studying ultrafast temporal evolutions, for instance the motion of photo-excited carriers in semiconductors, the optical pump probe method is a powerful technique. We can measure the evolution according to the time delay between the pump and the probe beams. Thus yielding information about the relaxation of electronic states in the sample. Therefore, we can compute the recovery time in the sample using the reflectivity changes.

Objective :

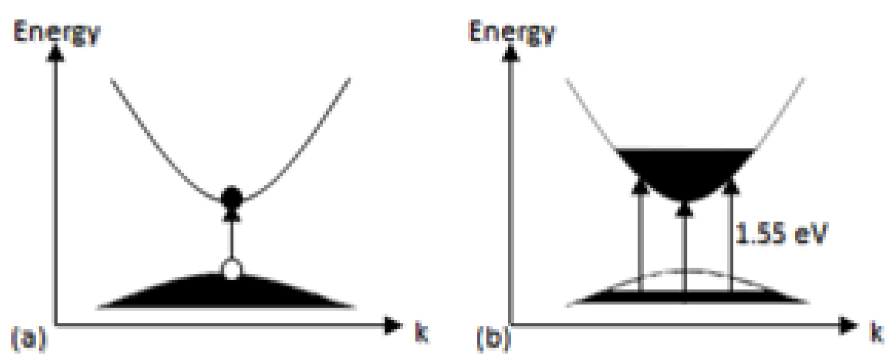
We monitor the intraband motion of carriers i.e. the electron-holes ensemble in the direct semi conductor GaAs depending on the excitation intensity.

Band Structure of GaAs



Walter, Cohen : Phys. Rev. (1969.07)

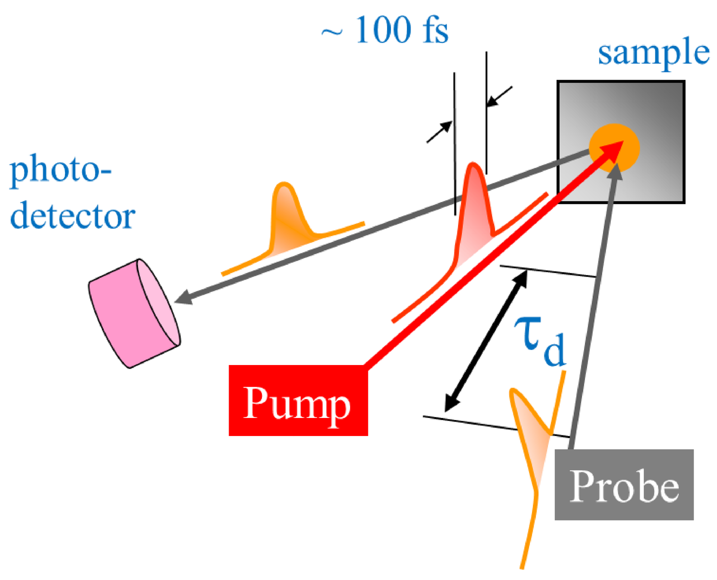
Band structures of GaAs



(a) Weak excitation. (b) Strong excitation.

Setup of the experiment :

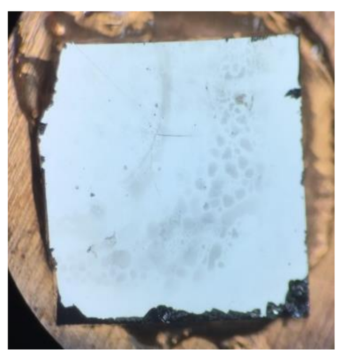
Pump probe method

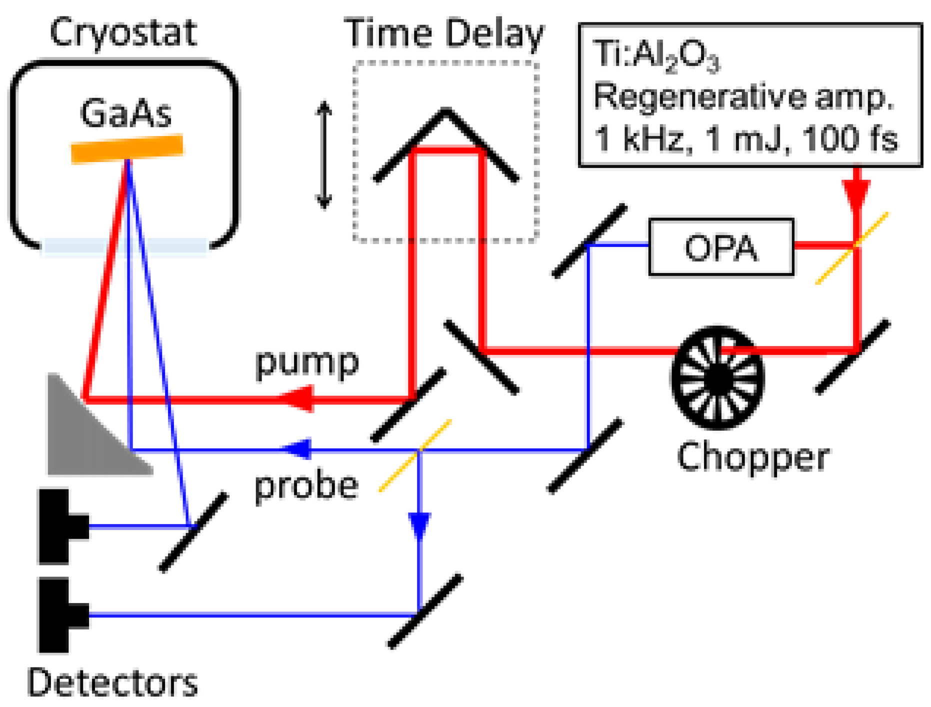


One pulse (the pump) will disturb the sample at t=0 while the other one will, after a delay, cross the sample and play the role of a probe.

Setup of the experiment.

GaAs Sample





Measuring ultrafast dynamics.

Ultrashort pulses.
Short time duration : keeps them from losing information, improves the time resolution.

We need a broad spectrum of frequencies in order to study the sample.

OPA (Optical Parametric Amplifier)
This process provides high output energies and a broad frequency tunability. During this experiment we will use mid-IR frequencies.

Results :

$$(1) \quad \frac{\Delta R(t)}{R} = Ae^{\frac{t}{\tau_1}}\left[1 - e^{\frac{t}{\tau_2}}\right]$$

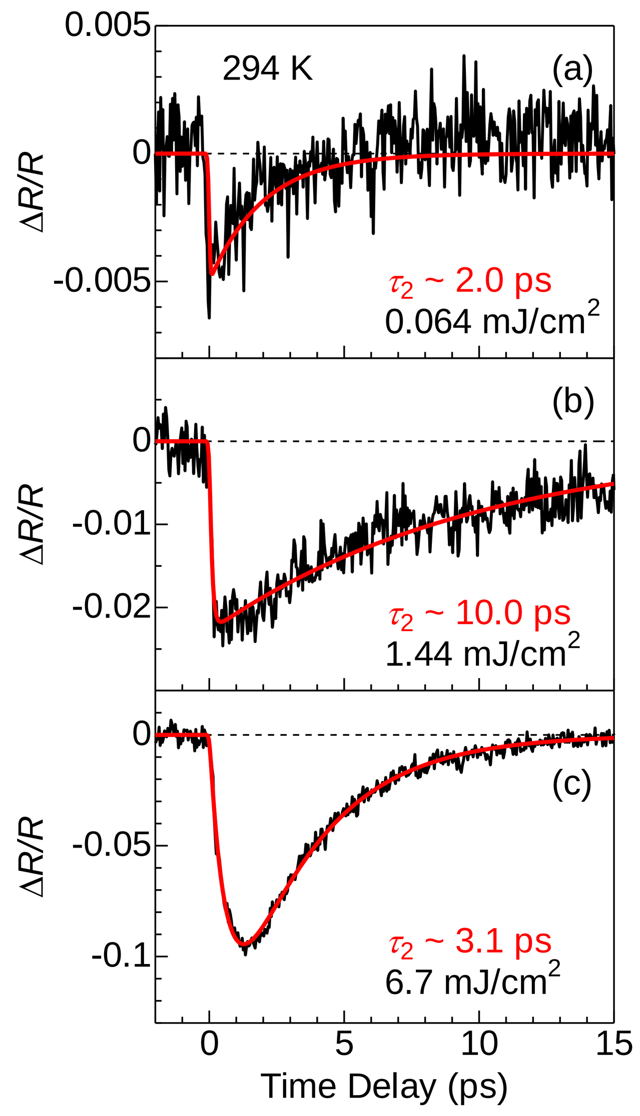
We proceed to the fitting of the exponential model (1) that allows us to determine precisely the decay time τ_2. $\frac{\Delta R(t)}{R}$ is the transient reflectivity, I_{ex} the excitation intensity.

In each case, the reflectivity decreases first, then grows back to its previous value.

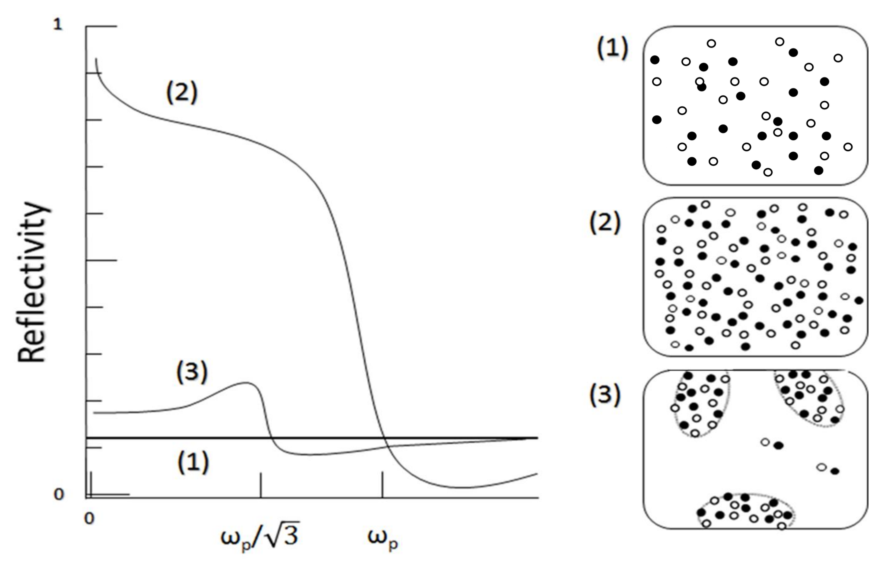
The evolution of the decay time isn't linear given the intensity. If beforehand τ_2 increases from 2.0 ps to 10.0 ps in parallel with the excitation density, it then decreases from 10.0 ps to 3.1 ps while the excitation density still increases.

I_{ex} (mJ/cm^2)	0.064	1.44	6.7
A	-0.005	-0.023	-0.18
Rise τ_1 (ps)	0	0	0.81
Decay τ_2 (ps)	2.0	10.0	3.1

(a)-(b)-(c) Temporal evolution of reflectivity change ΔR/R in GaAs.



Explanation :



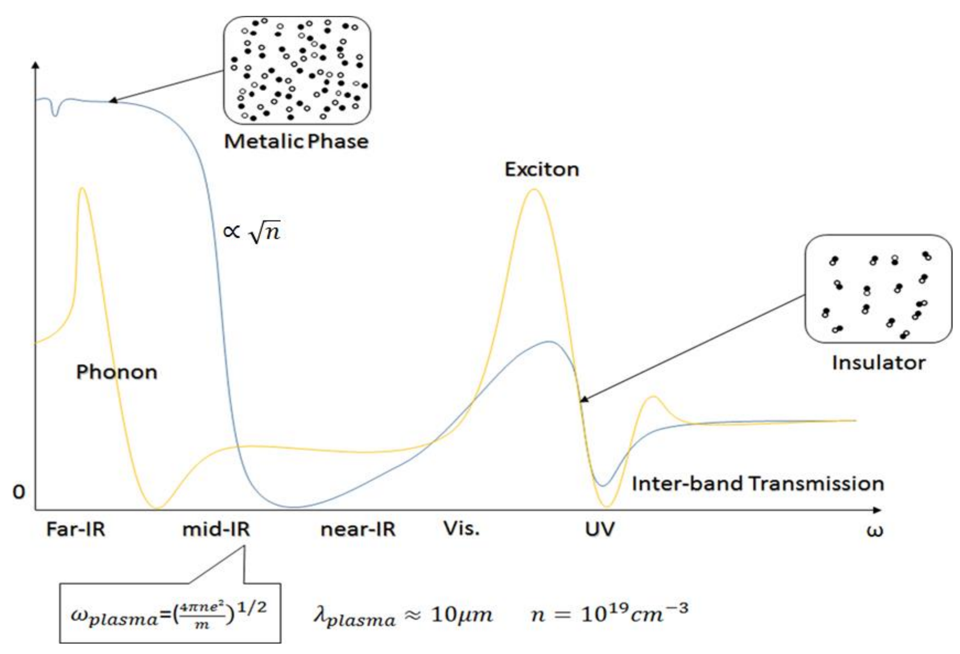
Nagai, Shimano, Kuwata-Gonokami : Phys. Rev. Lett. (2001.06)

GaAs is a direct gap semiconductor, intrinsically the radiative recombination prevails and causes the fast reduction of the plasma resonance at the surface.
The dynamic change in our results reflects the modification of the carriers motions. Our experiment takes place at room temperature.
This figure shows the reflectivity around the plasma frequency $\hbar\omega_p$ at room temperature for different situations of electron-hole system.

At weak excitations (1), we have small amount of free carriers which are all stuck into impurities at once.

The carrier density exceeds the amount of impurities (2). The electron-hole system turns then slowly into a homogenous plasma.

In (3) we have a metallic state with Electron Hole Droplets (EHD), collective carriers behaving differently than the plasma, explaining the changes in the decay time.



Kuwata-Gonokami : Surikagaku (SAIENSU-SHA, 2004.10)

This figure shows the electromagnetic response of the high density electron-hole system, we observe that a strong metallic reflection appears in the mid-IR region. We would like to study this phenomena in the future.

Summary :

We measured the reflectivity changes in GaAs at several excitation densities using the pump probe method.
In the future, we would like to extend to the mid-IR and examine the transient reflectivity spectra to explain the drop of the metallic phase using the dielectric constant ϵ_b of a semi-conductor as follows : $\epsilon_m = \epsilon_b\left(1 - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}\right)$ where $\omega_p = \sqrt{4\pi ne^2/\epsilon_b m}$ is the plasma frequency, n, m, and e are the carriers' effective density, mass and electric charge. γ is the plasma damping.