L.2: Ultrafast carrier dynamics in metal nanoplatelets

Ultrafast transient transmission measurements have been carried out at the Laser Physics Application Division of RRCAT to study the carrier dynamics in metal nanoparticles.

The sensitivity of the detection of carrier dynamics can be enhanced if there is a surface plasmon resonance (SPR) at the photon frequency. For the photon frequencies near the inter-band absorption, the carrier dynamics results from a combination of intra and interband absorptions. This makes it difficult to isolate the pure intra-band dynamics in nanoparticles. To isolate the intra-band dynamics silver nanoplatelet colloids have been prepared, having the SPR peak at 1.59 eV, which is far from the interband edge of Ag. The transmission (T) spectrum of a sample is shown in Fig.L.2.1.

For the time-resolved dynamics studies, a Ti:Sapphire laser with 100fs pulse duration, 82 MHz pulse rep-rate, operating at 780 nm wavelength, was used. The pump-probe transmission set-up is shown in Fig.L.2.2. The mechanical chopping of the pump, and detection using a lock-in amplifier, allows one to measure $\Delta T/T$ as small as 10^{-5}. Fig.L.2.3 shows the plot of $\Delta T/T$ for three different pump powers. For the lowest pump power of 10 mW, the decay is found to be exponential with a decay constant of 710 fs. However, as the pump power is increased, the transmission recovery becomes slower.

To understand this behavior, one looks at what happens when the pump pulse is absorbed by the electrons in the nanoparticles. Due to this absorption, the electron distribution in the p-band of silver gets modified, leading to the initial fast rise in the transmission. At the laser powers used in the present work, the electron temperatures can reach up to few thousands of Kelvin. The lattice temperature, however, still remains close to the room temperature, in the initial stages. As the time progress, the electron-phonon interaction leads to cooling of the electron and heating of the lattice. The rate of the heat transfer, from the electrons to the lattice, depends on the specific heat capacity of the electrons, which, in turn, depends on the temperature. This leads to the observed increase of the decay time with increasing pump power. Calculations of electron temperature evolution based on the above model show the same dependence on pump power. Thus, the ultrafast optical transmission measurements provide a direct look at the heat transport in nanoparticles. This is an important parameter for designing devices using such nanoparticles.

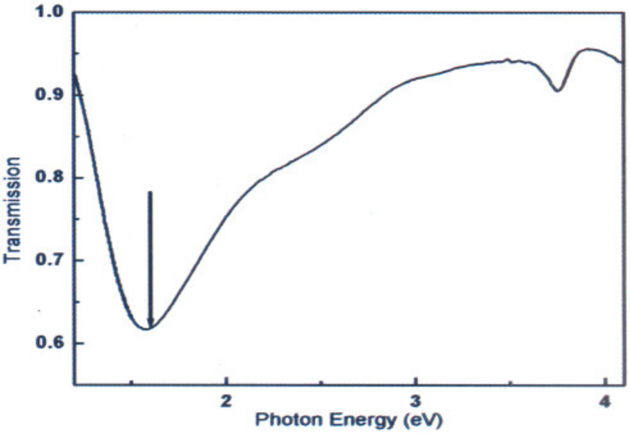

Fig.L.2.1:. The transmission spectrum of the silver nanoplatelet solution. The arrow shows the pump laser energy.

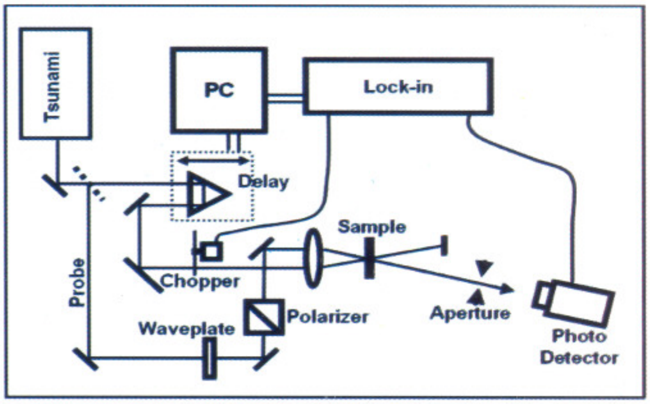

Fig.L.2.2: The pump-probe set-up used for the measurement of the ultra-fast changes in the transmission of the silver nanoplatelets.

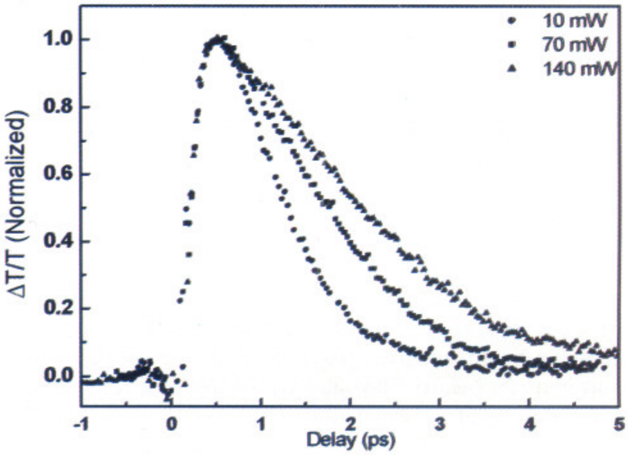

Fig L.2.3: The transient changes in the transmission of the sample for different pump powers.

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