Time resolved mid-infrared absorption in silica: ultrafast heat transfer observed by direct probing of anharmonic vibrations

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Precise control of laser material structuring requires a comprehensive understanding of the light-matter interaction at the ultrafast timescale. This understanding includes the electronic processes governing the absorption of laser energy and the consequent relaxation, through electronic or vibrational pathways, driving to structural changes. In this framework, attention was paid to exploiting the potential of mid-infrared radiation (MIR) interacting with the crystal lattice, through active infrared vibrational modes for material manipulation [1].
Here, we propose a novel approach based on the selective probing of vibrational modes in the MIR to investigate the electron-phonon coupling timescale in non-linear excitation conditions in bulk glassy materials. Indeed, thanks to the possibility to drive the irradiated region out of the equilibrium state, with sub-picosecond time resolution, it will be possible to monitor fundamental mechanisms underlying light-matter interaction in strong fields, exploring the vibrational activation through the direct observation of the phonon dynamics. The experimental investigations were conducted on amorphous silica, as model material for transparent dielectrics. To explore the characteristic time for electron-lattice energy exchange, we investigated the Si-O-Si antisymmetric stretching overtone localized at approximately 4415 nm. This feature was chosen due to its relevance in the main structural changes of silica glass and its sensitivity to the distribution of the Si–O–Si bond angle. Furthermore, by probing an overtone, our measurements will be particularly sensitive to the anharmonicity of the atomic potential, allowing us to follow charge-induced potential distortions.
The overtone dynamics was followed using an ultrashort probe beam (pulse duration 200 fs, centered at 4450 nm) excited with a pump pulse of 50 fs (centered at 800 nm), operating at intensities near the optical breakdown threshold. Monitoring the rapid temporal evolution of the transient absorption (TA) dynamics, it will be possible to follow the lattice response as a function of the pump-probe delay (Figure 1).

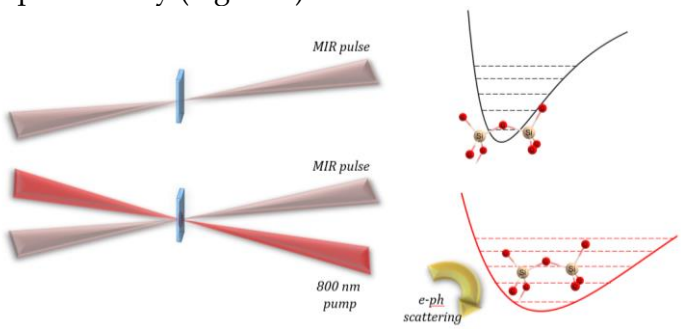

Figure 1. Phenomenology of the experiment.

Our results emphasize the potential of this method, which can be easily extended to other local markers of vibrational coupling within the host glass structure. This underscores the efficacy of our approach as a powerful tool to investigate various dynamics, such as electron-phonon coupling, vibrational excitation transfer, solid-solid phase transitions, 2D infrared spectroscopy and more.

References:
[1] M. Först et al., Nature Phys 7, 854–856 (2011).
[2] P. Daguzan et al., Phys. Rev. B 52, 17099 (1995).