# Controlling Material Transformation and Plasma Emission with Trains of Ultrafast Laser Pulses

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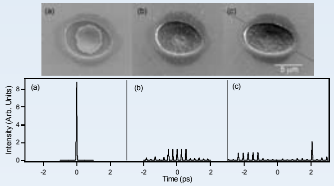
## Introduction

The sequence of events terminating in the ablation or desorption of material from the surface of a solid can be highly complex. Nevertheless, if the initial excitation step is coherent, it may be possible to exploit this coherence to control the subsequent behavior of the solid. This concept is especially relevant for semiconductors, where phonon excitation plays an important role. Our motivation is to use a sequence of ultrafast laser pulses to drive the optical phonons of a crystal, and to monitor the subsequent modification of the surface. The idea is to set the spacing of the pulses to a multiple of the optical phonon period so as to induce wide amplitude motion of the lattice vibrations.

### SEM images of GaAs craters:



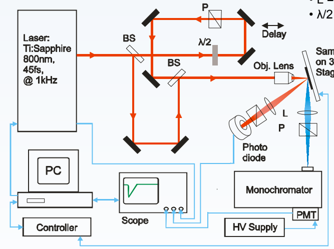
- The molten phase produced by single pulse is largely absent for the 5-pulse train and completely absent for the 5+1 train.

We tried to understand the energy transfer within the solid, mass and energy transport into a plume above the surface, plasma formation, and interaction of the plasma with the surface and with the radiation field upon irradiation of pulse train. In this study, the simple pulse trains, a double pulse and three pulses, are used. Significant differences of double pulse ablation compared with single pulse were observed in ps time scale. The result was even more surprising in the sub ps time scale when a three-pulse train was used with the variable spacing between the pulses.

## Double pulse ablatio study:

### Experimental set up

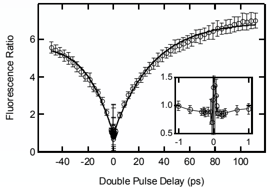
- P = polarizer
- BS = beam splitter
- L = lens
- λ/2 = half wave plate



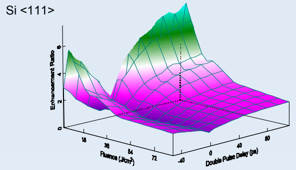
- The laser beam is focused to about 3.6 μm in diameter.

## Results

### Fluorescence experiments on Si<111>



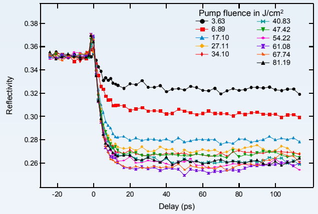
- Ionic fluorescence of Si<111> vs. delay time at a double pulse fluence of 10.40 J/cm$^2$.



The enhancement ratio, E(F,t) or the ratio of the fluorescence signal for the pulse pair to that for a single pulse having the same total fluence (F), is plotted as a function of delay time (t) and F.

- E(F,t) always increases with |t| for a fixed F.
- At a fixed delay E(F,t) decreases with fluence.

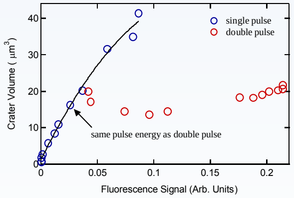
### Reflectance study using 800nm pump and 400nm probe:



- The reflectance change of Si <111> surface after the pump pulse. The reflectance increases first to a higher value, because of the greater reflectivity of the induced plasma.[1] The reflectance subsequently decreases to a low value because of surface damage.

[1] C.V.Shank *et al.*, *Phys. Rev. Lett.* **50**, 454 (1983).

### AFM measurements of the craters:



- The fluence for single pulse craters ranges from 6.2 to 91.4 J/cm$^2$.
- The fluence for double pulse is 41.5 J/cm$^2$, with the delay ranging from 0 to 105 ps.
- The volumes of the craters made by single and double pulse using same total energy were similar, even though the fluorescence increased significantly with delay.

### Si<111> Craters etched with 44% KOH solution:

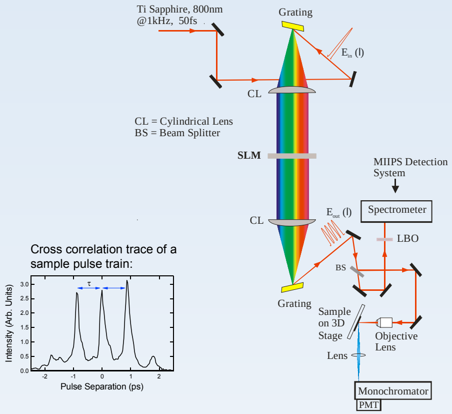
Double pulse delay: 1.5 ps 66 ps 86 ps

- The craters with longer delays have more molten material inside.[2]

[2] Z. Hu *et al.*, *Appl. Phys. Lett.* **90**, 131910 (2007).

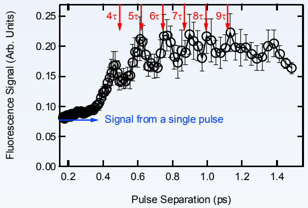
## Controlling LO phonons of GaAs using pulse train with variable delay:

### Experimental set up:



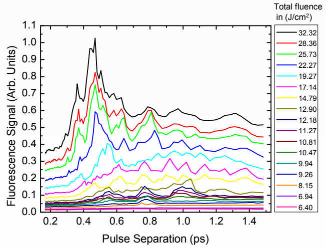
Cross correlation trace of a sample pulse train:

### Fluorescence experiments on GaAs:



- Fluorescence from the background continuum vs. spacing between the pulses at a three-pulse fluence of 14.79 J/cm$^2$. The peaks appear when the spacing of the pulses match with the integer multiple of the LO phonon period of GaAs.

- No such oscillations were observed for the sharp atomic lines (at 403nm and 417nm).

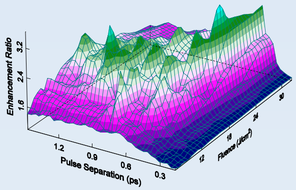


Fluorescence vs. pulse separation at different laser fluences.

- LO phonon oscillations are visible only for a certain fluence range e.g. 9.26 to 17.14 J/cm$^2$.

- At low fluence region the oscillations are very weak.

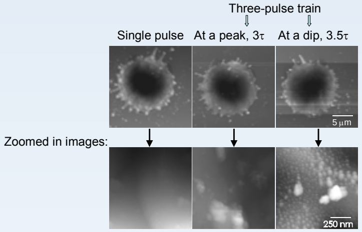
- At high fluence region the oscillations become chaotic and a new broad peak appears around 470fs.



The enhancement ratio is plotted as a function of delay time and laser fluence.

- The LO phonon oscillations are highly sensitive to the laser fluence.

### AFM measurements of the craters:



The laser fluence used for these craters is 13.59J/cm$^2$.

- In the first panel the morphology of the craters looked similar for the single and multi pulses.
- Zoomed-in images show nicely organized nanoparticles inside the crater for the dip which were absent for the peak.

## Conclusion

### Double pulse ablation:

- The first pulse superheats the Si surface and melts it electronically. The second pulse couples more strongly to the liquid phase, producing a larger fraction of electronically excited Si and Si$^+$, causing the enhanced fluorescence.

- The rise time of E(F,t) is determined by the propagation rate of the liquid phase into the solid.

- The AFM images of the etched craters show that the double pulse enhances the amount of material transformed.

- The plume produced by the first pulse partially shields the surface from the second pulse, causing E(F,t) to decrease with increasing F at fixed t.

### Control of GaAs LO phonons:

- The control of the background continuum and lack of control of the atomic/ionic lines suggest that the process is an electron recombination process, either free-free (Bremsstrahlung) or free-bound type.

- The laser energy and spacing of the pulse train plays an important role for the oscillation of the surface phonons and the signal enhancement is observed only at the resonant condition.

- The absence of nanoparticles at the resonant pulse separation suggests large surface oscillations which destroys the distribution of the nanoparticles.

## Acknowledgement

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