# Ultrafast Bandgap Photonics: Meta-stability of Transient States.

Michael K. Rafailov
University of Alberta,
Edmonton, Canada T6G 2V4

## ABSTRACT

Ultrashort high intensity pulse creates extreme non-equilibrium condition in bandgap material producing dramatic perturbations in electronic structure that, in its turn, leading to changes in electronic, magnetic, and optical states of condensed matter. Interesting experimental results have recently been reported on transient phenomena ranging from ultrafast laser induced detection denial to light induced high temperature superconductivity. While single pulse interaction with bandgap material is well observed, explained and documented, one of the major problem is to maintain meta-stability of such matter states: stability of transient effects that may last well beyond thermalization time. The objective of this paper is to discuss approaches to meta-stability of transient states.

**Keywords:** ultrafast bandgap photonics**,** light induced transient effects, meta-stability of transient states, dynamics of collective states, correlated electron systems, light-induced phase transition, light-induced superconductivity.

## *1.* INTRODUCTION

Ultrashort pulse (ultrafast) laser interactions with matter has been shifting our views on matter for more than three decades. Ultrashort high intensity laser pulse produces dramatic perturbations in electronic structure that may lead to fundamental changes matter states.

Transient optical, electronic and magnetic effects induced in condensed matter by high intensity ultrashort pulse have been described, verified and some theoretical models explaining the effects have been established [1-6]. Transient cooperative changes in condensed matter states under the exposure of ultra-short laser pulse became a focus of Ultrafast Bandgap Photonics and some experiments have demonstrated fascinating results like light induced high temperature superconductivity [7].

Transient processes induced by high intensity but low energy pulses - like ones generated by ultrafast lasers, are remarkably distinctive from other types of light-matter interactions because of significance of athermal component. With extreme intensity in single ultrashort laser pulse energy simply may not be enough to raise temperature in bandgap material via single pulse. Therefore phonon-related energy transfer phenomena may be reduced to minimum and thermal effects can be weak in single-pulse interaction-either with low energy photons or with low phonon rate. Even more, thermal component may be kept in control while high intensity high frequency but low energy pulse train is interacting with bandgap material. That, in its turn means that low energy ultrashort pulse interaction with bandgap material by no means could lead to lattice temperature raise.

Ultrashort high intensity-low energy pulse interaction phenomena may be considered as pure transient effects if sufficient energy build-up is not occur in matter. Timing of transient effects may vary from few picoseconds to few nanoseconds. Transient is the key-word in such athermal process and maintaining meta-stable conditions by preserving transient state is one of the greatest challenges for Ultrafast Bandgap Photonics. Meta-stability of transient states is leading to great practical applications not only in Photonics but generally in applied physics. Applications of such remarkable discoveries as light induced high temperature superconductivity, light induced bandgap engineering, light induced transparency, light induced magnetism are just only few of many prospective research topics where maintaining meta-stability of transient states can lead to great applications.

Paper is focused at meta-stability of transient states therefore describes ways of transient state extention well beyond of single pulse induced transient timing. Meta-stability of transient states in the paper is focused mostly at phenomena that have been the scope of papers presented in April 2016 at Ultrafast Bandgap Photonics Conference in Baltimore MD.

*rafailov@ualberta.ca

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## 2. ENERGY TRANSFER WITH ULTRASHORT PULSE

While transient absorption/transmission induced by light - that is commonly known as bleaching, have been in spot light of short pulse laser-induced transient effects for quite a while and have been exploited in many scientific and industrial applications, it also provided deep inside view on ultra-short pulse energy transfer into bandgap material that has been a subject of studies in bleaching-based pump-and probe experiments.

Basically, there are two competing modes of energy transfer from ultrashort pulse: athermal and thermal modes. With different type of pump-and-probe experiments the timing of energy transfers have been thorough investigated and energy levels corresponding with the progress of athermal and thermal modes have been defined. Athermal mode is preceding the thermal one and defined by rapid changes in energy aka temperature of excited carriers. With the time scale starting from few picoseconds that mode precedes thermal mode where phonons dominate energy transfer. That mode may last as long as nano-seconds and eventually leads to temperature changes in the material. Figure 1 depicts timing of energy transfer as well as direct observation of transmission changes as a result of ultrafast pulse energy transfer as well as mechanisms involved in energy transfers and their timing.

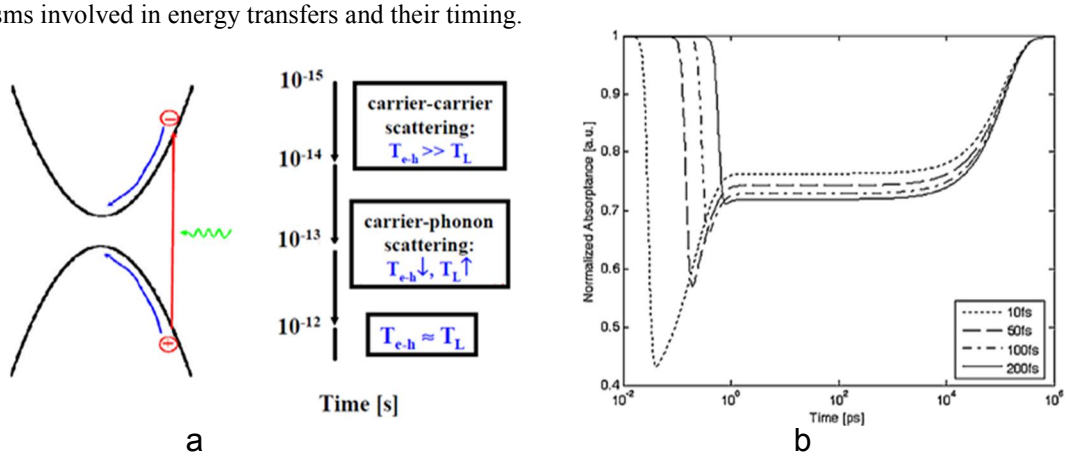



Figure 1. Energy transfer in bandgaps. a. timing of energy transfer; b. direct observations of Si bleaching in near-IR with different pulse width[8].

There are multiple back-to back processes initiated by ultra-short pulse in bandgap material but generally those processes are lasting relatively short time in athermal and much longer – in thermal mode when lattice involves into interaction with energy transitioned via carrier - phonons scattering. All effects discussed by Ultrafast Bandgap Photonics are related to perturbations and dynamic changes related to excited electrons: those electrons that traditionally called "hot electrons" and those that co-exist with so called "cold lattice". To keep the lattice "cold" is a major challenge for Ultrafast Bandgap Photonics.

. There are long term conditions when energy deposited via ultrashort pulses can "sink" without leaving thermal footprints in the material. In such a way ultra-short pulse train interacting with bandgap material may extend lifetime of light-induced transition effects as well as to reduce required pulse energy and respective power consumption. Way to do it is to "engineer" of ultra-short laser pulse train. Pulse train engineering is intended for not allowing the thermal mode energy deposition to exceed a threshold that minimizes or even eliminates transient athermal effects–and not only for the time of single pulse interaction, but for much longer time-again, in the presence of a mechanism that prevents or minimizes energy build-up into lattice. Therefore, at some point two competing mechanisms of energy transition are coexists; one– via emission or directed carrier motion, and another one via carrier-phonon lattice interaction that builds up energy deposition into lattice. With controlled portion of energy deposited by phonons into lattice the latest may be considered as transient athermal effects as well leading to some vibrational changes in the lattice [7].

It is possible to maintain meta-stable state of transient effects with carefully modulated pulse train envelope intensity and variable duty cycle therefore preventing accumulation of energy in the lattice and, therefore, preventing the development of long lasted thermal effects in the material.

Due to *Heisenberg principle* photon excitation happens in the continuum of energies exceeding the energy that is necessary for direct transition - bandgap energy. Excited carriers are generated in such a way that they possess an excess of energy determined by the difference between energies of continuum of photon excited carriers and very specific band

gap energy - ($h\nu - E_g$). Carrier-carrier interaction leads to equalization of free carrier energies inside of the both bands: free electrons in the conduction band and holes (vacancies) in the valence band – see Figure 1, a. However, energy-equalized continuum of free carriers still has effective temperature $T_e$ significantly higher than lattice temperature $T_L$. Therefore, the next step in energy transition is energy transfer into the lattice via carrier-phonon interaction. Important comment - if at that point excited carriers are removed – whatever via emission or by directed motion with an external field, e.g., as in detection, then carrier-phonon transition may be diminished.

Energy transferred from an electron to the lattice during single interaction is just a fraction of total excessive energy, therefore full energy re-distribution in the conduction band requires multiple interaction in order to reach energy balance inside the band. That time of energy relaxation –thermalization, is the time that require the system to return to equilibrium state.

Thermalization or energy-redistribution may last much longer than excitation and energy re-distribution in hot electrons –measured in Ge and InSb that energy redistribution time is in few picoseconds [8, 9, 10]. Pump-probe technique has been used to investigate processes in solids and an actual change in the lattice has been detected with either X-ray diffraction on lattice [9, 10] or by measurement of transient absorption of a sample [8]. Lattice changes measured by X-ray diffraction peak scatter have very specific time scale and timing that is not depending on transferred amount of energy. Direct observations of lattice changes following femto-second optical excitations were reported with X-ray diffraction measurements [9, 10] and, those results shows that lattice changes start in few pico-seconds after pumping laser pulse.

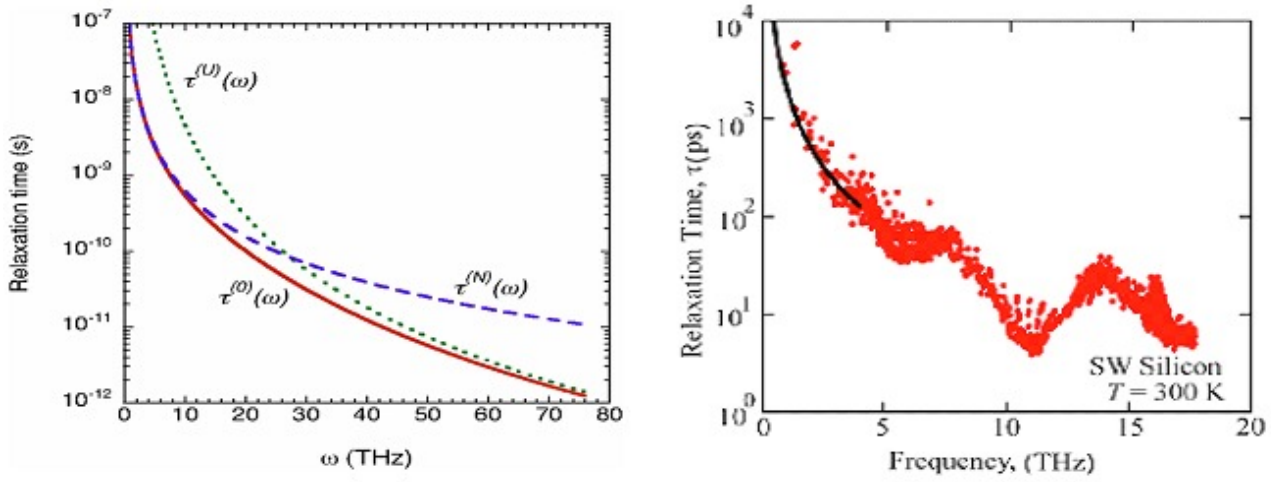
Light-induced transitional absorption is direct result of *Pauli blocking* resulting of *Pauli principle* that leads to depletion of valence band electrons. Direct answer for time scale changes in semiconductor optical characteristics provides transmittance changes measurements after the excitation by ultra-short pulse. Observations of lattice changes following femto-second optical excitations were reported with relative optical transparency measurements [9] and, those results shows that full thermalization process is lasting from 1.7 to 2 pico-seconds as well. Transient time with pumping in IR for other important semiconductors are: GaAs – 7ps; for Si -1ps; Ge-1.1 ps and InSb – 1.7ps.

## 3. EXCITATION

The whole idea of meta-stability of transient states is to keep non-equilibrium state that is originally lasting from few picoseconds for much longer period of time by maintaining the concentration of "hot" electrons that define that transient state but keeping lattice "cold". Normally such conditions can coexists only for short period of time after a pulse, and if energy accumulated by the lattice is exceeding a threshold then transient state eventually disappears with lattice temperature raise. That threshold is defined by specific concentration of excited carriers that depends on lattice temperature and a transient state collapses when the concentration is depleted below a level necessary for maintaining the state at given temperature, which is- in its turn, rises due to energy deposition into the lattice.

Two conditions need to be maintained for meta-stability of a transient state– conditions that seem to be mutually exclusive at first glance – to limit energy transition via phonons and therefore preventing lattice temperature increase, while maintaining high concentration of hot electrons that is necessary for supporting any transient state. To limit energy transition via phonons we need to provide a way for energy to "sink" out of material sample after electron-electron interaction: such as particular material sample morphology –size and dimensions, like strips of thin films, and/or with applying an external field directing excited carriers out, or with an emission. In the same time we need to maintain necessary concentration of excited carriers and, therefore a sample needed to be pumped periodically with laser pulses to maintain excited carrier concentration that is necessary for the existence of a transient state. That periodic excitation shall keep excited carrier concentration up to a level that supports transient effects. With high energy photons full relaxation time is higher while electron-to-electron energy transfer is pretty much on femtosecond scale, therefore, to maintain meta-stability we need to pump with fast rate and it increases energy deposition into the sample that may rapidly increase lattice temperature.

One way of doing it is to arrange pumping in relatively low energy regions - like, Mid-to Long Wave IR or even THz regions, and to pump a sample with a repetition rate that is up to thermalization time, e.g. with multi-GHz and THz pulse repetition rate. Figure 2 – after [11] demonstrates relaxation time plot versus pumping energies.




a. b.

Figure 2**.** Frequency-related phonon relaxation time in bandgap material [11]: a. calculations, where normal (dashed curve), wide-angle (dotted curve), and total (solid curve) relaxation times plotted as a function of frequency for Si at *T*=300 K.; b. experiment.

With higher frequency and, therefore higher pumping photon energy thermalization time decreases reaching in IR single digit picoseconds, such as in the mentioned experiments. From one side it makes easy to keep up necessary concentration of excited carriers, from another it increases energy that has no way to dissipate but heats the lattice that may lead to disappearance-collapse of the transient effects.

From that point pumping in mid-IR/THz region- in the region between 10THz and 20THz, makes it easy to maintain meta-stability and, not allowing a sample to accumulate heat and keeping lattice "cold" along with high concentration of excited carriers. Pulse repetition rate for maintaining meta-stability shall be in tens of GHz.

The time scale of observable meta-stable transient effects is highly dependable on pulse energy, repetition rate and pulse widths. That allows pulse trains with easy achievable ultrashort pulse repetition rate – with respect to existing capabilities of ultrafast lasers "comfortable" rate might be well below 50GHz – see Figure 3 (after [12,13], where such lasers can manage sufficient energy per pulse with respect to pulse width along with necessary pulse repetition rate.

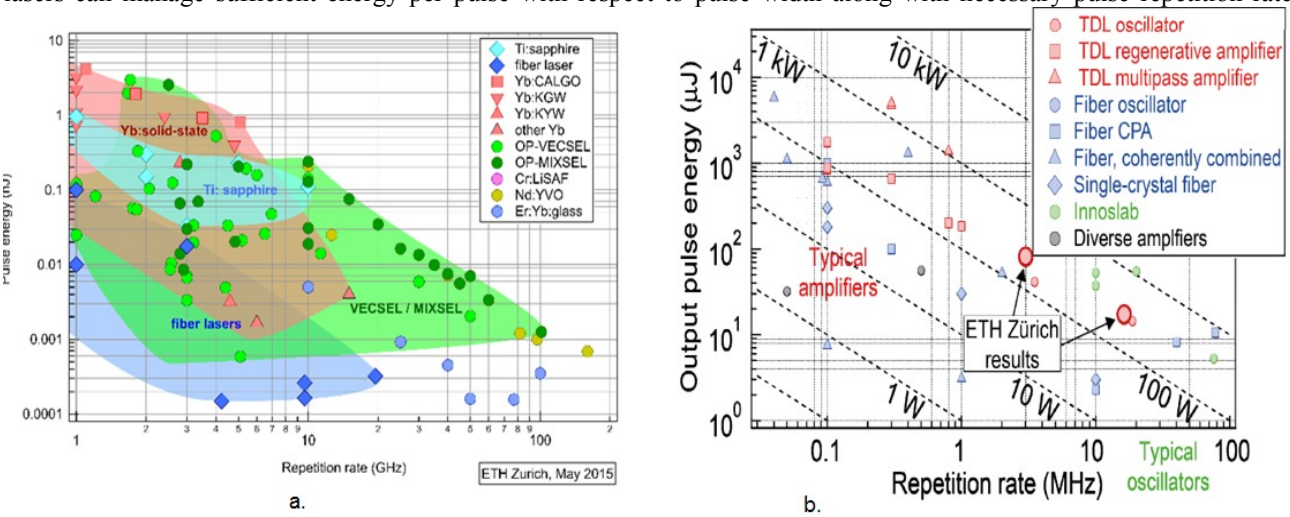



Figure 3. Ultrafast lasers available for pumping. a. nJ per pulse in multi-GHz pulse repetition rate [12]; b. μJ per pulse in multi-MHz pulse repetition rate [13].

## 4. CONCLUSIONS

Meta-stability of transient states can be achieved with periodic pumping of a sample with ultrafast laser pulses. Meta-stability based on maintaining of necessary excited carriers concentration but preventing temperature raise in a sample. With carrier-to carrier scatter pretty much independent of photon energy the pumping pulses repetition rate depends on lattice-carrier thermalization time. And thermalization time depends on pumping wavelength: with shorter wavelength the relaxation time decreases. However, higher photon energy increases sample temperature disproportionally, that leads to collapse of a transient state.

Going after lower energy pumping photons may eliminate lattice temperature increase and, in the same time will not require high pumping pulse repetition rate but there is some inconvenience of such ultrashort laser pulses generation in THz region. However, in a "sweet spot", that is located around 15THz or 20 micron, the pulse repetition rate in tens of GHz may work well for maintaining meta-stability of transient states-with respect to sample bandgap, sample geometries and sizes as well as energy removal mechanisms. Going deeper into THz region may be beneficial – due to lower energies, and with respect to maintaining excited carrier concentration may require lower pulse repetition rate, but it still in GHz.. Going to shorter wavelengths, like to mid-IR and near-IR may require deep modulation of pulse train intensity with pulse repetition rate close to THz..

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