Optically driven attosecond electron dynamics in gallium arsenide

Fabian Schlaepfer[1], **Matteo Lucchini**[1], **Shunsuke A. Sato**[2], **Mikhail Volkov**[1], **Lamia Kasmi**[1], **Nadja Hartmann**[1], **Angel Rubio**[2], **Lukas Gallmann**[1], **Ursula Keller**[1]

1. Physics Department, ETH Zurich, 8093 Zurich, Switzerland

2. Theory Department, Max Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany

A fundamental understanding of ultrafast electron dynamics in solids induced by light is of great interest for future high-speed electro-optical devices operating in the petahertz frequency regime [1]. In the last years, a number of publications demonstrated the possibility to resolve and control carrier dynamics in semiconductors [2,3] and dielectrics [4,5] on the few- to sub-femtosecond time scale using attosecond transient absorption spectroscopy (ATAS). These experiments were performed with a *non-resonant* pump pulse, i.e., pump photon energies smaller than the corresponding band gap. Here in contrast, we resolve for the first time the attosecond carrier dynamics induced by a *resonant* intense laser pulse. We study the attosecond electronic response in gallium arsenide (GaAs), a technologically important narrow band gap semiconductor [6].

During the light-matter interaction, an infrared (IR) pump pulse can induce two types of processes in the semiconductor: *inter*- and *intra*-band transitions. Their distinct roles in light-driven electron dynamics are highly debated and still not fully resolved. We present a detailed study investigating the dominating transition in the transient response and discuss the influence of the non-linear interplay between the two mechanisms on the ultrafast electron injection from the valence (VB) into the conduction band (CB).

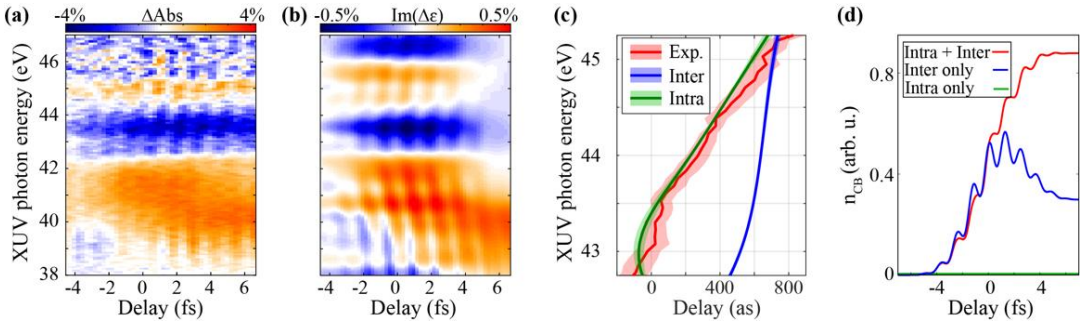

Fig. 1 (a), (b) Measured and simulated transient absorption modulations induced by an intense *resonant* pulse. **(c)** Energy-dependent delay between the pump field intensity and transient absorption oscillations extracted from the measured signal and the simulations with only *intra*- or *inter*-band transitions included, respectively. **(d)** Simulated number of electrons excited from the VB into the CB for the different limits.

In our setup, we combine an intense few-cycle IR pump pulse with an attosecond extreme-ultraviolet (XUV) probe pulse. The IR can induce *intra*- and *inter*-band transitions, while the subsequent XUV pulse probes the modified carrier population around the band gap via electron injection from the As-3d core level. As shown in Fig. 1(a), the pump pulse induces a complex delay- and energy-dependent structure of enhanced (red) and decreased (blue) XUV absorption regions. Transient oscillations appear over a broad energy window.

With state-of-the-art first-principles calculations, we can reproduce the measured features with very good agreement (Fig. 1(b)). To understand the physical mechanism responsible for the transient signal, we use a simplified three-band model, which allows to study the influence of *intra*- and *inter*-band transitions separately. By comparing the oscillation phase of the transient signal in the different limits (Fig. 1(c)), we can conclude that the attosecond dynamics induced by the IR pulse is dominated by *intra*-band motion, rather than by *inter*-band transitions. Given the *resonant* pump condition, this is an unexpected finding.

Furthermore, we study the influence of the interplay between the two processes on the injection mechanism of electrons from the VB into the CB (Fig. 1(d)). Interestingly, we find that *intra*-band motion assists the carrier injection induced by the *inter*-band transition significantly, eventhough it can by itself only accelerate carriers within single bands. A detailed theoretical study explains the enhancement of the number of excited photocarriers with an additional excitation pathway involving the *intra*-band motion [7].

Our findings represent an important step forward in understanding the petahertz electron dynamics in semiconductors, relevant for future ultrafast opto-electronic devices.

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35 word abstract

We present measurements of attosecond electron dynamics in single-crystalline gallium arsenide induced by a resonant optical light pulse. Our results, supported by ab-initio simulations, demonstrate a complex electronic response involving intra- and inter-band transitions.