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Valley-resolved Electronic Coherences in Silicon Observed by Attosecond Transient Absorption Spectroscopy

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Abstract: Electronic coherences are observed in silicon by attosecond transient absorption spectroscopy. Various sub-4 fs oscillations across the conduction band reveal complex couplings between valence-conduction and conduction-conduction bands indicating pathways for coherent preparation of highly excited electrons.

I. INTRODUCTION

Understanding the absorption of light and subsequent carrier dynamics in semiconductors plays a crucial role for optimizing next-generation photonic devices for increasingly faster performance. However, a direct access of dynamics during carrier excitation for a multitude of possible band transitions when using broadband light remains challenging.

Here, attosecond transient absorption spectroscopy is employed for studying band couplings, i.e. electronic coherences, in single crystalline silicon during excitation by an intense 5-fs optical pulse (1×10^{13} W/cm²).

II. RESULTS & CONCLUSION

Transient absorption changes in the conduction band (CB) of silicon are monitored by an attosecond pulse at the silicon $L_{2,3}$-edge (~99.8 eV). The recorded transient (Fig. 1a) features a multitude of oscillations across the CB. In a frequency-over-energy Fourier analysis in comparison to the band structure, couplings can be identified by lines with unit slope (Fig. 1b & c). Besides observing 2ω oscillations consistent with previous observations of the NIR field driving electrons across the band gap, the data suggests that the optical pulse can coherently couple the valence band (VB) and CB at the L and Γ points by a multiphoton process (black dashed lines and circles). However, some newly observed features, can be assigned to CB-CB coherences (purple dashed lines), notably between the L_1/L_3, $\Gamma_{15}/\Gamma_{2'}$ and K_3/K_1 critical points, with K_3/K_1 requiring two photons. A possible path for creating CB-CB coherences can be understood by the leading edge of the pulse first indirectly exciting carriers into the Δ_1 valley via the indirect gap excitation and subsequently the main pulse initiating the coupling among the conduction band density of states. The time domain measurement allows measuring lifetimes of these coherences as well as their sequence of generation. Detailed time-domain analysis and supporting TDSE simulations will be presented.

In conclusion, the results provide insight into complex couplings between bands that take place during excitation with broadband ultrashort laser pulses, an effect that should be general for most semiconductor materials. Specifically, couplings between CBs expose pathways for generating highly excited electrons in semiconductors. Monitoring electronic coherences valley-resolved opens prospects for control of hot carrier generation by electric field engineering.

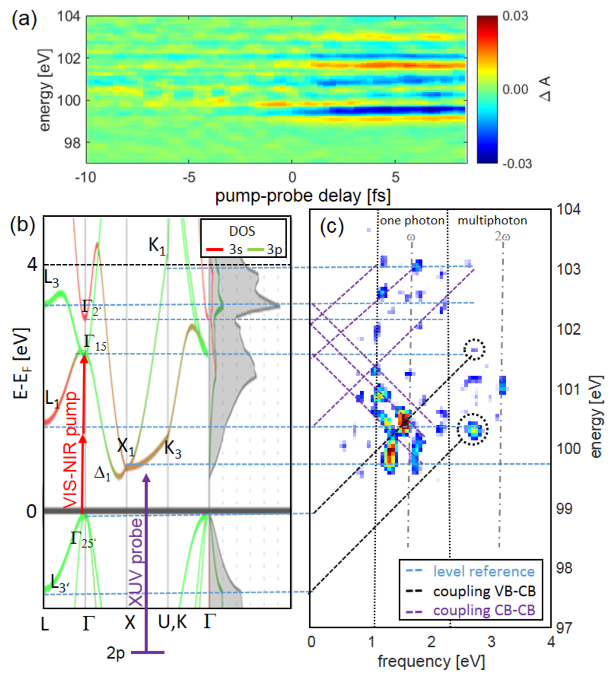

Fig. 1. (a) Transient absorption changes at the silicon $L_{2,3}$-edge. (b) Si band structure and density of states (DOS) indicating critical points and the pump-probe scheme. (c) Fourier analysis of (a) revealing various VB-CB and CB-CB electronic coherences. Only one possible excitation pathway is indicated (b) for clarity.

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