The role of crystal orientation in attosecond photoinjection dynamics in germanium

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Abstract. Ultrashort light pulses can be used to manipulate electronic and optical properties of solids at extreme temporal scales, paving the way to the study of ultrafast electron dynamics. In recent years, attosecond-based spectroscopic techniques have proved to be an instrumental tool in such studies. To this end, we investigated the effects of crystal orientation on ultrafast photoinjection dynamics in germanium using attosecond transient reflectance spectroscopy (ATRS) aided by time-dependent density functional theory (TD-DFT) calculations. Our results show that ATRS is sensitive to subtle changes in the transient reflectance due to crystal orientation, although carrier photoinjection in germanium is qualitatively robust against crystal rotation, displaying similar photoinjection processes and timings at two different crystal angles.

1 Introduction

During the last decades, laser technology has evolved in such a way to allow us to generate light pulses with shorter and shorter duration, down to the attosecond (10^{-18} s) level. This gave us access to previously unobserved properties of solid-state materials, for instance through the study of light-matter interaction at time scales comparable with the motion of electrons in solids, with enticing applications in fields like optoelectronics [1]. For this reason, Attosecond Transient Reflection and Absorption Spectroscopy (ATRS and ATAS, respectively) have been widely employed to investigate ultrafast phenomena like the photoinjection of carriers from valence (VB) to conduction (CB) band in a semiconductor by means of intense, few-cycle, near-infrared (NIR) pulses. Since numerous mechanisms like single- or multi-photon absorption, band-dressing, tunneling excitation, or intra-band motion [2] take place during nonlinear light-matter interaction, their comprehensive physical description is hard to achieve. Nonetheless, Inzani and coworkers [3] successfully described these phenomena in monocrystalline, undoped germanium, a narrow-gap semiconductor, by combining ATRS measurements to a double-theoretical approach.
In this work, we use a similar strategy to investigate the role of crystal orientation on ultrafast photoinjection dynamics in germanium. Our results not only demonstrate that NIR-induced photoinjection dynamics in Ge are robust to crystal orientation, displaying the same evolution in time and being controlled by the same mechanisms, but also the validity of ATRS as a technique to probe the role of lattice orientation on the observed dynamics.

2 Experimental setup

Germanium possesses a zincblende crystal structure, that is, a face-centred cubic lattice with a basis composed of two germanium atoms in (0,0,0) and $(\frac{1}{4}a, \frac{1}{4}a, \frac{1}{4}a)$, with lattice constant a = 5.66 Å. We decided to study the crystal when the pump laser field is oriented along the [010] and [110] directions. These two configurations are obtained by rotating the sample along the [001] axis by $\frac{\pi}{4}$ and are expected to differ the most. A further rotation of $\frac{\pi}{4}$ would indeed bring the NIR field aligned with the [100] direction, formally identical to the [010]. The NIR pump - centred around 1.55 eV, lasting ~5.3 fs and with an intensity of 10 TW/cm^2 in vacuum (1.2 TW/cm^2 inside the crystal) – and XUV probe - spanning 25–45 eV (thus covering the $M_{4,5}$ absorption edges at 29.2 eV and 29.8 eV) – impinge onto the sample at an angle of 66° (Fig.1).

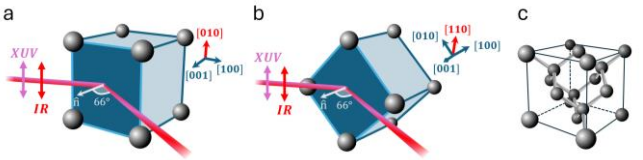

Fig. 1. Scheme of the experiment showing the two sample orientations, [010] (**a**) and [110] (**b**), and the polarisation of the IR and XUV pulses with respect to the Ge crystal unit cell. (**c**) Crystal structure of germanium.

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3 TD-DFT simulations

The first step of our research was performing theoretical simulations through TD-DFT, to gauge the differences between the two crystal angles in terms of charge distribution in the k-space. If we observe the residual electron population after the pump pulse is over (10 fs after its peak), we notice that in the [010] configuration the electrons appear uniformly distributed along the four diagonals of the k-space (Fig. 2a). On the other hand, the [110] case presents a significant excitation mainly in two diagonals in the $k_y = -k_x$ plane (Fig. 2b), demonstrating that carrier excitation can be steered towards certain regions of the electronic band structure through crystal rotation. The temporal evolution of the total excited populations in the two cases was evaluated as well (the dashed lines in Fig. 3a represent the gauge-invariant time evolution, *i.e.* when $A_{IR}(t) = 0$). An in-depth analysis shows that in both cases the mechanisms at play and their relative timings are identical, with single-photon processes taking place mostly on the pulse rising edge, and tunnel ionisation and multi-photon processes acting towards the pulse peak, with oscillations at twice the IR frequency caused by intra-band motion (see Ref. [3]). The final total population is mainly due to multi-photon absorption, with the [110] case having a slightly (5%) higher total population.

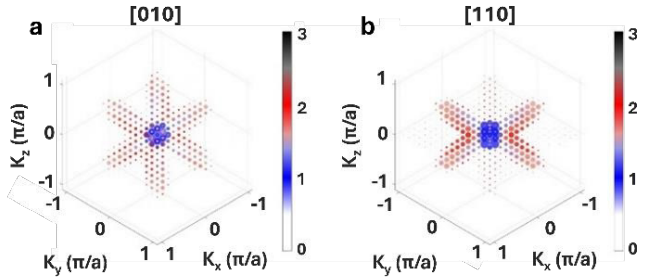

Fig. 2. Excited population in Ge 10 fs after the IR peak in the [010] (**a**) and [110] case (**b**). Blue, red, and black spots represent single-, two-, and three-photon transitions respectively, and their size is proportional to the population in that specific k-point.

4 ATRS measurements

ATRS measurements were subsequently performed to further investigate the picture suggested by the theoretical predictions. We constructed a differential reflectivity trace by acquiring the reflected XUV spectra both in presence (I_{on}) and in absence (I_{off}) of the IR pump in rapid alternation, subtracting the latter from the former and normalising to I_{off}, thus obtaining $\Delta R/R = (I_{on} - I_{off})/I_{off}$. The delay between pump and probe was changed at steps of 0.33 fs, and the energy resolution was 20 meV around Fermi energy (E_F = 29.3 eV). We performed two sets on measurements, one in [010] and one in [110] configuration, and the results (Fig. 4) demonstrate that shape, position and temporal evolution of the transient optical features are similar one to the other, both displaying a few-fs build-up after the pump envelope and oscillations at twice the IR frequency caused by intra-band motion. On the other hand, the overall amplitude of the transient signals of the two configurations differs, being larger for the [110] orientation, as may be clearly seen in Fig. 3b. If we integrate our trace between the energies of 26.2 eV and 32.2 eV, where we have a stronger effect of real charges injected into the CB, we find that the evolution in time in the two cases is similar as well (solid lines in Fig. 3a), suggesting a possible higher photoinjection in [110]. This finding correlates well with the estimated number of injected electrons as calculated from TD-DFT.

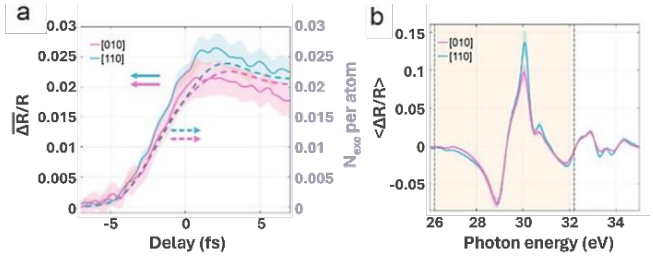

Fig. 3. (**a**) Solid curves: temporal evolution of the modulus of $\Delta R/R$ in the area marked in yellow in (b). Dashed lines: TD-DFT results for the time evolution of the excited electron population in CB. (**b**) Delay-averaged ATRS traces for the [110] case (cyan) and [010] case (magenta). Same colour coding as in (a).

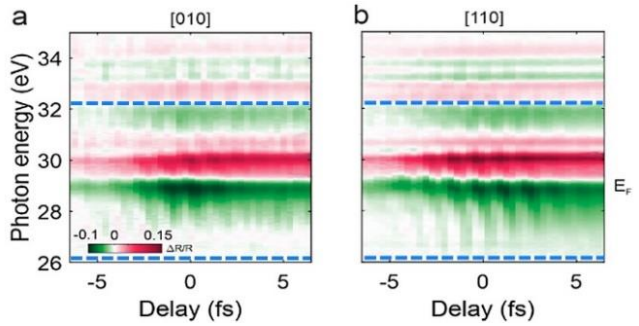

Fig. 4. Differential reflectivity traces with a crystal orientation of [010] in (**a**) and [110] in (**b**). A red (green) colour indicates a positive (negative) change in the sample reflectivity. The dashed blue lines indicate the extremes of the yellow region of Fig. 3b.

5 Conclusions

In this slice of our work, we showed through TD-DFT calculations that photoinjection by a few-fs, intense, NIR pulse in germanium is qualitatively robust against crystal orientation with a few key differences. Namely, different distribution of the populated k-points in reciprocal space and a slight increase of the total excited residual charge after interaction for the [110] case. We also performed ATRS measurements for the two orientations, revealing qualitatively similar features for both orientations. By further analysing the traces around Fermi energy we observed an increased strength of transient optical features in the [110] case and a time evolution which is close to that of the electron population in the CB.

We have therefore proven the capability of this technique to detect small differences in photoinjection processes, while also indicating crystal orientation as a further degree of freedom to control the momentum distribution of the excited electrons and their precise timing.

References

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