

Electric dipole oscillation in solids characterized by Fourier transform extreme ultraviolet attosecond spectroscopy

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Abstract. We characterized electronic dipole oscillations in chromium doped sapphire (Cr:Al₂O₃) using Fourier transform extreme ultraviolet attosecond spectroscopy (FTXUV) combined with an isolated attosecond pulse, which reveals the electric band-structure and dephasing process in solids.

1 Introduction

In recent years, an attosecond (as: 10⁻¹⁸ s) light source in the extreme ultraviolet (XUV) region has been developed [1,2]. The properties of extremely short wavelength and temporal duration will open the way to monitoring ultrafast electric motion [3,4]. Here, we study the electric dipole oscillations in chromium doped sapphire (Cr:Al₂O₃) characterized by Fourier transform extreme ultraviolet attosecond spectroscopy (FTXUV) combined with an isolated attosecond pulse (IAP) [5].

2 Experiment

Figure 1(a) shows a schematic view of the setup for FTXUV based on transient absorption spectroscopy. The IAP and near-infrared (NIR) pulse were focused on the Cr:Al₂O₃ (36-nm thick) without substrate. Figure 1(b) shows the energy level diagram of the Al₂O₃ [6]. In this experiment, the Al₂O₃ was doped with the Cr material during the single-crystalline α -Al₂O₃ crystal growth. The Cr³⁺ ions produced a donor-like intermediate state for the Al₂O₃ host material. The Al₂O₃ host material had the bandgap energy of 8.7 eV, while the Urbach tail region corresponding to the intermediate state was 5 to 8.7 eV. The target intensity of the NIR was 2×10¹² W/cm². The intense NIR pulse induced an interband polarization from the valence band (VB) state to intermediate and conduction band (CB) states.

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Simultaneously, the IAP with high photon energy (44-eV center photon energy with 192-as duration [2]) allowed the excitation of electrons in the VB, intermediate, and CB states. In this pump-probe system, the timing jitter was 23 as at the root mean square over 12 hours [4].

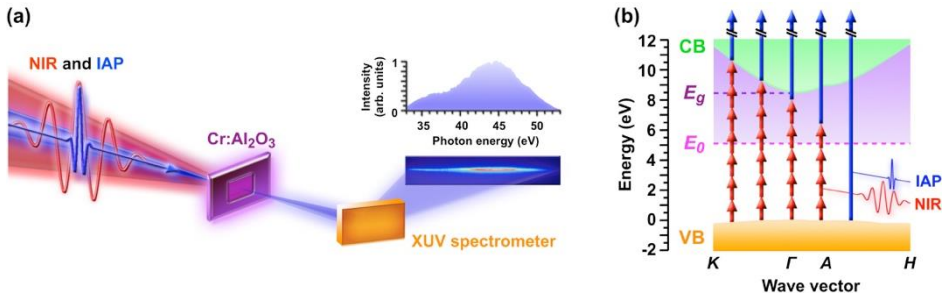


Fig. 1. Experimental setup and energy level diagram of Cr:Al₂O₃. (a) Schematic view of the FTXUV. IAP: 192-as duration in the Cr:Al₂O₃. NIR pulse: 1.6-eV, 7-fs duration. The targets were Cr:Al₂O₃ (36-nm-thick) without substrate. The transmitted IAP from the target was detected by the XUV spectrometer. (b) Energy level diagrams of Cr:Al₂O₃. The orange, purple, and green shaded areas show the VB, intermediate, and CB states. Bandgap energies E_g was 8.7 eV in the Al₂O₃ host material. The initial intermediate state E_0 with Cr dopant was located at 5.0 eV. The blue and red arrows show the IAP and NIR, respectively.

3 Results and discussion

Figure 2(a) shows the transient absorption trace of Cr:Al₂O₃. The observed characteristic temporal modulation on the trace indicates the dipole oscillations in Cr:Al₂O₃. The coherent broadband IAP excites the electrons in the VB, intermediate, and CB states to a high-level CB state (quasi-continuum state). The induced electronic state leads to quantum interference, resulting in a temporal modulation of the IAP absorption spectrum. Figure 2(b) shows the energy components after Fourier transformation of the temporal delay axis in the bottom trace of Fig. 2(a). The intense NIR pulse (2×10^{12} W/cm²) produces electron

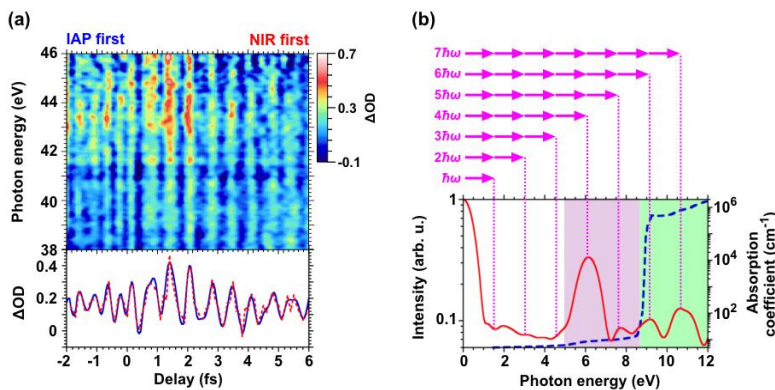


Fig. 2. FTXUV trace and energy component. (a) The transient absorption trace in Cr: Al₂O₃. The trace shows the deviation of optical density (ΔOD) with and without the NIR pulse. The bottom trace shows the integrated line profiles (red dashed line) for the whole energy region in the upper graph. The blue solid line is the interpolation process line profile. (b) The line profile spectra (red solid line) after Fourier transformation for the temporal delay axis in (a). The pink dotted lines correspond to the photon energies of the NIR pulse. The $\hbar\omega$ corresponds to ~ 1.6 eV. The blue dashed lines show the linear absorption coefficient of Cr:Al₂O₃. The purple and green shaded areas correspond to the intermediate and CB states, respectively.

transitions corresponding to $4\hbar\omega$ energy components. The $6\hbar\omega$ and $7\hbar\omega$ energy components are higher than the CB of 8.7 eV (green shaded area). The $4\hbar\omega$ and $5\hbar\omega$ energy components correspond to the intermediate state (purple shaded area). These results indicate that the FTXUV combined with the IAP can identify the band structure through the ultrafast periodic electron motions.

Figure 3(a) shows the each line profile in Fig. 2(b) after the windowed Fourier transform. The intermediate components ($4\hbar\omega$ and $5\hbar\omega$) have a relative time delay of approximately 2 fs from the CB components ($6\hbar\omega$ and $7\hbar\omega$). For a better understanding of this time delay, we numerically simulated the resonant high-order polarizations using the model in a two-level system [7], which consists of the ground state and the excited state either resonant to the intermediate state ($5\hbar\omega$) or resonant to the CB state ($7\hbar\omega$). The calculated results with the parameter value of $\mu F/\hbar\omega=0.59$, where μ and F correspond to the strengths of the transition dipole moment and the applied NIR driving laser field (7-fs duration). Assuming that the CB state ($\tau_{CB}=0.2$ fs) has the faster dephasing time of 2.8 fs

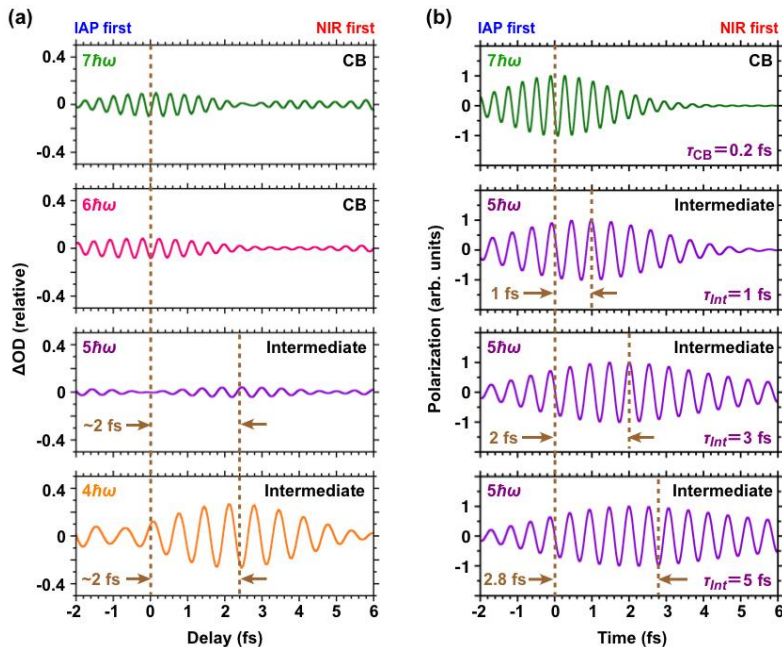


Fig. 3. Time-dependent dipole oscillation and polarization. (a) Measured dipole oscillation in Fig. 2(a) after windowed Fourier transform. The window energies of Fourier filtering are 6.2 ($4\hbar\omega$), 7.7 ($5\hbar\omega$), 9.2 ($6\hbar\omega$), and 10.8 eV ($7\hbar\omega$). The applied window energy width is ± 0.35 eV, taking into account the NIR bandwidth. The $4\hbar\omega$ and $5\hbar\omega$ correspond to the intermediate state. The $6\hbar\omega$ and $7\hbar\omega$ correspond to the CB state. The electric dipole oscillations with periodicities of 667 ($4\hbar\omega$), 537 ($5\hbar\omega$), 450 ($6\hbar\omega$), and 383 as ($7\hbar\omega$) correspond to frequencies of 1.5, 1.9, 2.2, and 2.6 PHz, respectively. (b) Calculated time-dependent polarization. Upper trace (purple solid line) shows the Fourier filtered polarization in the CB state ($7\hbar\omega$) with $\tau_{CB}=0.2$ fs. (d) CB resonances. Bottom three traces show Fourier filtered polarizations in the intermediate state ($5\hbar\omega$) with $\tau_{Int}=1$, $\tau_{Int}=3$, and $\tau_{Int}=5$ fs, respectively. The brown dotted lines and arrows show the time delay between the CB and intermediate components.

relative to the intermediate state ($\tau_{\text{int}}=3$ fs), the observed time delay of approximately 2 fs is reproduced well, as shown in Figs. 3(b). Generally, the dephasing time in the spatially localized energy state is much longer than that in the band energy state [8]. Thus, the spatially localized intermediate state with the low Cr doping level has many fewer relaxation channels in the unoccupied state compared with the CB state. Consequently, the delayed buildup of the polarization could be observed in the resonant polarization of the spatially localized intermediate state. In addition, the dephasing time is commonly explained by the density- and energy-dependent damping rate [9], which is proportional to cube root of the excited carrier density n ; the value of n depends on the fluence of pump pulse and the absorption coefficient of the target [10]. The linear absorption coefficient in the CB state drastically increases higher than the fourth order of magnitude compared with the intermediate state, as shown in Fig. 2(b). Thus, the excited carrier density in the CB state might be higher than the intermediate state even though the nonlinear multiphoton excitation. The different carrier densities could produce individual dephasing times in the intermediate and CB states. The tunable time delay induced by polarization behavior would be designed by band-structure control through the kind of dopant material and doping level. This tunability will further assist with the flexibility of the petahertz signal manipulation.

4 Conclusion

The characterized dipole oscillation and dephasing process using the FTXUV is useful for studying variety of electronic devices and laser crystals with doping technology. This study will lay the essential groundwork for exploring the condensed matters.

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