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Zooming Surface- and Junction-Sensitive Electron-Ionic Dynamics in State-Specific Tabletop Transient XUV Absorption/Reflection Spectroscopy

In recent decades, significant progress has been made in developing tabletop time-resolved X-ray spectroscopy and related studies, offering both precision in probing elements, carriers, and oxidation states, and flexibility for instrumental modifications. By utilizing gas-phase high-harmonic generation to produce attosecond pulse trains, extreme ultraviolet (XUV) probes have enabled the exploration of electronic localization dynamics with millielectronvolt resolution and high surface sensitivity, leveraging the optical transitions from shallow core levels. Typically, polarons are formed in transition metal oxides through the interactions with optical phonon baths, which are dominant in the adiabatic regime. The lattice reorganization energy in this case is so large that the first electron–optical phonon scattering event creates a small polaron without requiring extensive carrier thermalization. Using the transient XUV absorption/reflection spectroscopy, it was observed for the first time that disrupting the iron-centered octahedra in the rare-earth orthoferrite ErFeO3 leads to the formation of nonadiabatic polarons. Coherent charge hopping between neighboring Fe^{3+} and Fe^{2+} sites persists for several picoseconds before the polaron fully forms. The observed small polaron formation time is an order of magnitude longer than previous measurements, indicating a shallow potential well, even in the excited state. These findings highlight the importance of accounting for dynamic electronelectron correlations, along with electron-phonon–induced lattice changes, in understanding small polaron behavior for applications in transport, catalysis, and photoexcitation, which can be uniquely measured with the XUV probe.

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