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Experimental demonstration of attosecond pump–probe spectroscopy with an X-ray free-electron laser

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Pump–probe experiments with subfemtosecond resolution are the key to understanding electronic dynamics in quantum systems. Here we demonstrate the generation and control of subfemtosecond pulse pairs from a two-colour X-ray free-electron laser. By measuring the delay between the two pulses with an angular streaking diagnostic, we characterize the group velocity of the X-ray free-electron laser and show control of the pulse delay down to 270 as. We confirm the application of this technique to a pump–probe measurement in core-ionized para-aminophenol. These results reveal the ability to perform pump–probe experiments with subfemtosecond resolution and atomic site specificity.

Footnotes

This work is published as Guo, Z., Driver, T., et al. “Experimental demonstration of attosecond pump–probe spectroscopy with an X-ray free-electron laser.” *Nature Photonics* (2024): 1-7.

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