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标题: Water's contribution and enzyme's work - a KITA study

作者: Born, B (Born, Benjamin); Sagi, I (Sagi, Irit); Havenith, M (Havenith, Martina)

编者: Farkas DL; Nicolau DV; Leif RC

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摘要: Enzyme dynamics control and contribute to myriads of mostly well-characterized metabolic processes. Yet, understanding the role of the aqueous reaction matrix remains an experimental challenge. By kinetic THz absorption (KITA) spectroscopy, we have studied the dynamic interplay between water and a human metalloenzyme at work in real-time. In our KITA setup, we combined a THz-time domain spectrometer (THz-TDS) with a stopped-flow mixer to study reactions with millisecond time resolution. We used picosecond THz pulses which directly probe hydrogen bond formation and breaking in the water network to observe enzyme-water interactions upon enzyme catalysis at the active site of a matrix-metalloprotease. During formation of the productive Michaelis complex, we detected a perturbation of coupled enzyme-water network dynamics. Supplemented by real-time biophysical techniques and molecular dynamics simulations we characterized the enzyme-water interplay in the particular case of enzyme catalysis. Our results suggest a polarization-induced gradient of water dynamics at the remote active site of a metalloenzyme with decelerated hydration water dynamics towards the active site. The observed long-range gradient of collective water motions might facilitate productive binding of substrates to enzyme active sites. Further KITA experiments shall improve our understanding of water's contribution to biological function.

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地址: [Born, Benjamin; Havenith, Martina] Ruhr Univ Bochum, LS Phys Chem 2, D-44780 Bochum, Germany

通讯作者地址: Born, B (通讯作者), Ruhr Univ Bochum, LS Phys Chem 2, Univ Str 150, D-44780 Bochum, Germany

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