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Spectroscopic probes of enzyme-ligand interaction dynamics
CHRISTOPHER CHEATUM, JIGAR BANDARIA, SAMRAT DUTTA, SARAH HILL, AMNON KOHEN, University of Iowa, Department of Chemistry — Formate dehydrogenase catalyzes the NAD-dependent oxidation of formate to carbon dioxide. The intrinsic chemical step involves hydride transfer from formate to the nicotinamide ring of NAD. As with several other NAD-dependent dehydrogenases, kinetic measurements suggest that thermal fluctuations of the enzyme are important in the hydride-transfer reaction. We have measured the dynamics of enzyme-inhibitor interactions in binary and ternary complexes of formate dehydrogenase with pseudohalides using infrared photon-echo spectroscopy. The pseudohalides are excellent vibrational chromophores that are known to be sensitive reporters of interactions with their local environments. They are also excellent inhibitors for formate dehydrogenase. Our measurements reveal significant differences in the dynamics of the different binary and ternary complexes. By comparing and contrasting the dynamics for different complexes we gain insight into the active-site components that make the most important contributions to the observed dynamics.

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Christopher Cheatum
University of Iowa, Department of Chemistry

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