

Studying ultrafast photorelaxation in functional Fe(II) complexes

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The photodynamics of functional complexes of iron is nowadays in the forefront of ultrafast science. Complexes, such as $[\text{Fe}(\text{terpy})_2]^{2+}$ (terpy = 2,2':6',2''-terpyridine) with long high-spin state lifetime, are potential candidates for photo-switchable elements, while others, such as $[\text{Fe}(\text{bmip})_2]^{2+}$ (bmip = 2,6-bis(3-methyl-imidazole-1-ylidene)-pyridine) with long metal-to-ligand charge transfer state lifetime, could be utilized as photosensitizers.[1] Photorelaxation dynamics of these complexes are extensively studied by transient spectroscopies and time-resolved X-ray scattering. With the help of time-dependent density functional theory, we determined potential energy surfaces for these two systems and then performed wavepacket dynamics simulations on them via the Multiconfigurational Time-Dependent Hartree (MCTDH) method. In case of the $[\text{Fe}(\text{bmip})_2]^{2+}$ molecule, we simulated the photorelaxation dynamics by explicitly describing the interaction of the molecule with the pump laser field. We studied how the laser pulse parameters could influence the relaxation dynamics and how it should be reflected in the simulated transient x-ray scattering (TR-XRS) signal [2]. In case of $[\text{Fe}(\text{terpy})_2]^{2+}$, the wavepacket motion is simulated in the lowest quintet state after photorelaxation in order to rationalize the measured TR-XRS signal.

References

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