

Maintaining vibrational coherence with electron spin

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Electron spin is an important property that determines processes such as chemical reactivity and the lifetime of the electron state. Spin is exploited in several applications such as luminescent materials, phototherapy, photochemistry, solar energy conversion, for which inducing a long-lived high spin state is crucial.

When large molecules absorb light, their energy dissipates. This happens through several <u>electron states</u> that are characterized by a difference in electron configurations and spins.

In femtochemistry, vibrational coherence plays a crucial role. When exciting an assembly of molecules using a short pulse, from tens to few hundreds of femtoseconds, the molecules oscillate in phase at their characteristic vibrational frequencies. In this way, the response of the assembly of excited molecules is like that of a single molecule. Vibrational coherence is therefore an ideal way of tracking where and how the molecular configuration is at a given instant in time.

Transfer of vibrational coherence between <u>electronic states</u> of molecules has been reported since the early years of femtochemistry. However, none of these studies has ever involved <u>states</u> of different spins. The lab of Majed Chergui at EPFL within the Lausanne Centre for Ultrafast Science has now reported for the first time a transfer of vibrational coherence in the case of a diplatinum complex in solution.

The scientists used their advanced femtosecond transient absorption set-



up to follow step-by-step the transfer of vibrational coherence during a spin switch between the lowest two electronic states of the molecule.

The clear and unambiguous passage between these two states is even more remarkable considering that the solvent usually destroys coherence. The experimental results are supported by quantum mechanical simulations showing the importance of the solvent in driving and modifying pathways and efficiency of energy flow in polyatomic <u>molecules</u>.

"The solvent is not just a spectator in photobiology and photochemistry but it can strongly affect the outcome of a function or reaction. Understanding its role is crucial for our description of nature and for future applications," says Majed Chergui.

More information: Roberto Monni el al., "Vibrational coherence transfer in the ultrafast intersystem crossing of a diplatinum complex in solution," *PNAS* (2018).

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