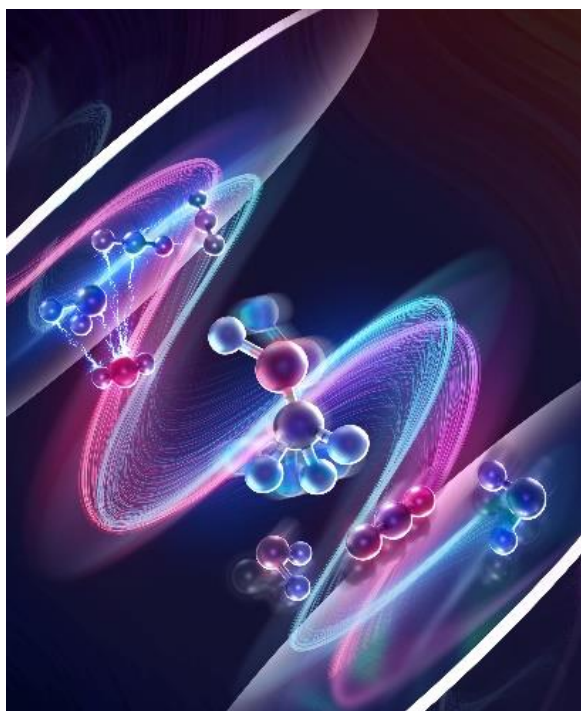


Ultrafast Dynamics of Molecular Polaritons

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<https://uni-due.zoom-x.de/j/64228670246?pwd=RjVQeFNIUkRKRkpiNVpKYXhJaFNldz09>



When the vibrational modes of molecules strongly couple with virtual states of photonic modes, they give rise to new states called molecular vibrational polaritons, accompanied by a significant population of dark reservoir modes. Similar to how atomic orbitals combine to form bonding and antibonding molecular orbitals in molecular bonding, polaritons resemble these orbitals, while the dark modes are akin to nonbonding orbitals. As polariton states are a hybrid of matter and light, their energy is shifted from the original states. This unique characteristic of polaritons leads to predictions that they can induce changes in chemistry under thermally activated conditions, giving rise to an exciting and emerging field called polariton chemistry, which has the potential to revolutionize the field of chemistry. However, despite some published results supporting this concept, the precise chemical physics and mechanisms underlying polariton chemistry are still not fully understood.

My group recently used 2D IR spectroscopy to address the mechanistic challenge by differentiate the dynamics of polaritons and dark modes. Specifically, our findings are twofold. Firstly, we discovered that polaritons can facilitate the transfer of vibrational energy within and between molecules. This paves the way for controlling the flow of vibrational energy in liquid-phase molecular systems. Secondly, by studying a single-step isomerization event, we confirmed that polaritons indeed have the ability to modify the dynamics of chemical reactions under conditions of strong coupling. In contrast, the dark modes behave similarly to uncoupled molecules and do not influence the reaction dynamics. This discovery affirms the central concept of polariton chemistry: polaritons can alter the potential energy landscape of reactions. Moreover, it provides a clear understanding of the role played by dark modes, establishing a critical foundation for the design of cavities in future polariton chemistry endeavors.

