

Where is the "action" in action-detected 2D spectroscopy?

A new perspective in The Journal of Chemical Physics, led by ICFO researchers, lays out the theory, methods and open challenges of ultrafast spectroscopies that read out what a material actually does with light: generate a current, emit light, and drive chemistry.

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How does energy move inside materials designed to harvest light, such as photovoltaic polymers, perovskites, photosynthetic complexes or atomically thin semiconductors? Over the past two decades, two-dimensional electronic spectroscopy (2DES) has become the method of choice to answer this question.

2DES correlates the colour of light a material absorbs with the colour at which it responds, femtoseconds to picoseconds later. This reveals which electronic states are coupled and how energy flows between them.

But energy materials are not built just to absorb light. They are built to do something with it: generate a current, emit light, drive chemistry. Conventional 2DES detects the optical field

radiated by the sample, so it cannot tell whether a given energy pathway actually contributes to a given function.

Action-detected 2DES (A-2DES) takes a different route. It measures the final product directly: photocurrent, fluorescence, emitted photoelectrons or ions, even tiny forces sensed by an atomic force microscope.

This opens new possibilities. Because the signal comes from the excited population itself rather than from a light beam emitted in a specific direction, A-2DES can be performed on a working solar cell, on a single molecule, or through a microscope. The technique can thus map ultrafast dynamics with spatial resolution down to the nanoscale. The detected product also selects which processes are seen: on a solar cell, the experiment reports on the excitations that end up as current.

But there is a catch. Measuring populations instead of optical fields changes the rules of the game. The experiment requires a sequence of four laser pulses with precise control over their timing and phase. The measured signal (for example, the photocurrent or the fluorescence) also contains additional quantum pathways that can partially cancel. In extended systems, another complication appears: excitations created independently in different parts of the sample can meet and interact after excitation. This produces an i¿½incoherent mixingi ?½ background that can dominate the spectrum and mimic genuine energy transfer even if i is

ot. ICFO researchers **Dr. Luca Bolzonello**, **Dr. Edoardo Amarotti**, **Dr. Joseph Wragg**, and **ICREA Prof. Niek van Hulst** have now published a perspective article on A-2DES in The Journal of Chemical Physics that brings together 18 researchers from Europe and the US, working across theory, instrumentation and applications.

The perspective builds **a common theoretical framework**, starting from simple model systems and moving up to large molecular aggregates, and quantifies when incoherent mixing takes over.

It then **reviews the strategies developed to isolate the meaningful signal**, from clever pulse orderings and polarization schemes to time-gated detection. It also compares detection modalities and smarter sampling strategies that cut measurement time.

Finally, it **discusses applications** to organic and perovskite solar cells, light-harvesting complexes, semiconductor monolayers, carbon nanotube devices and single molecules.

The central message is that, in action spectroscopy, the choice of observable is not a technical detail but part of the physics: what you detect determines what you see.

i¿½Action detection asks a different question than conventional 2D spectroscopy: not only how energy moves, but where it ends up,i¿½ says Dr. Luca Bolzonello, first author of the article. i¿½The two approaches are complementary, and together they give a much more complete picture of how energy materials actually work.i¿½

Reference:

Luca Bolzonello, et. al.; Action-detected two-dimensional electronic spectroscopy: Theory, methods, and future directions. J. Chem. Phys. 28 October 2026; 165 (16): 164202. https://doi.org/10.1063/5.0351936

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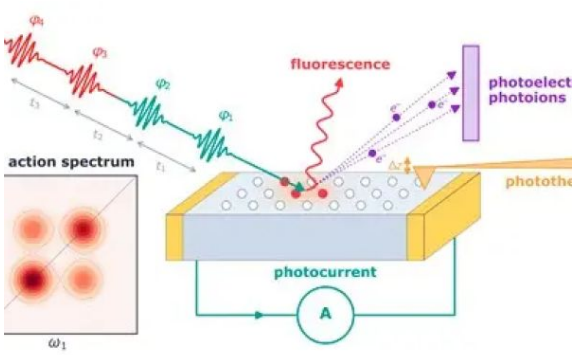

Scheme of A-2DES. Source: The Journal of Chemical Physics.