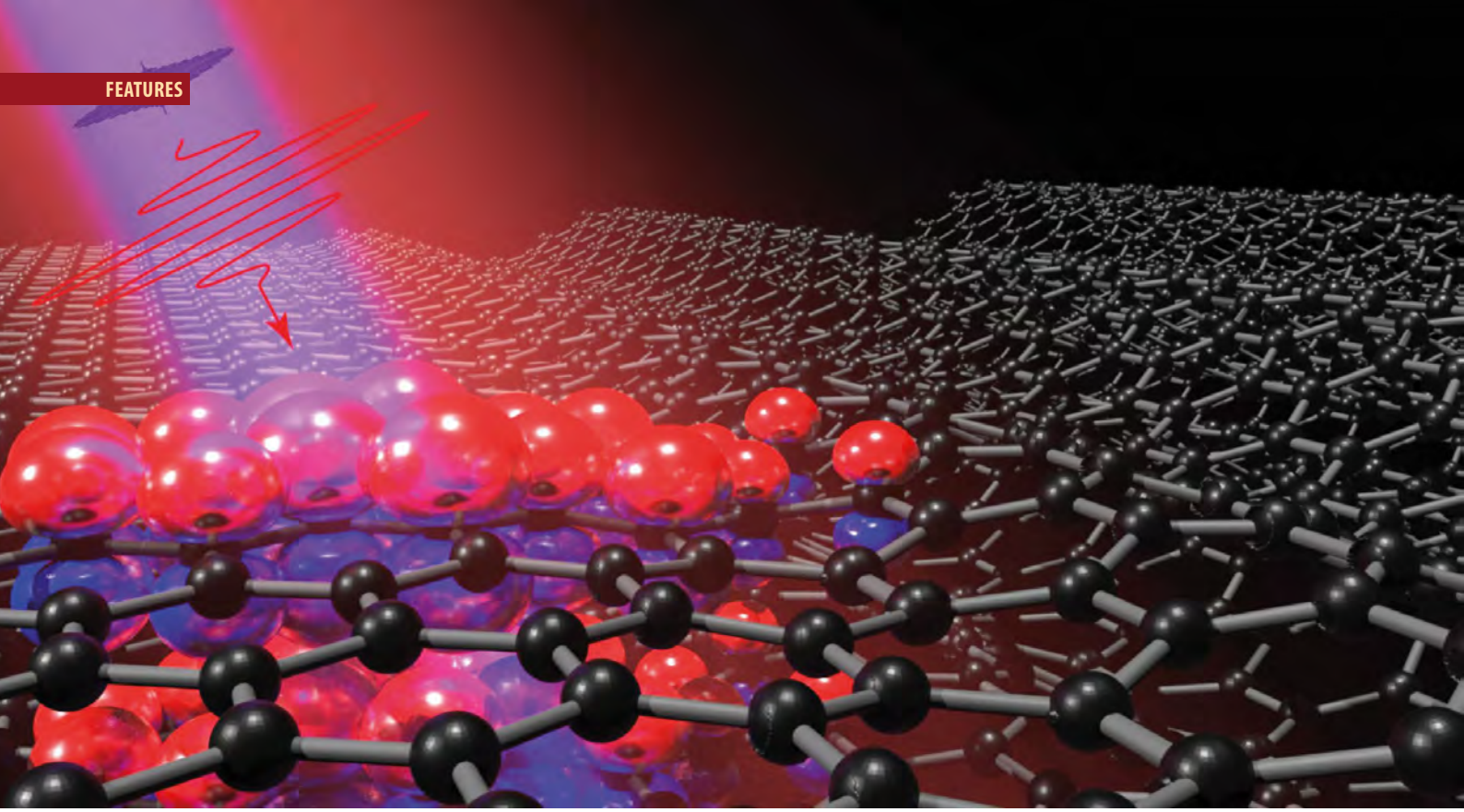


ATTOSECOND SCIENCE: A NEW ERA FOR MANY-BODY PHYSICS

Jens Biegert – DOI: https://doi.org/10.1051/epn/2024105
Attoscience and Ultrafast Optics, ICFO – Institut de Ciencies Fotoniques, The Barcelona Institute of Science and Technology, Castelldefels (Barcelona), 08860, Spain
ICREA - Institucio Catalana de Recerca i Estudis Avancats, Barcelona, Spain

The properties and the functionality of materials and devices, or chemical reactions, are determined by the microscopic interaction of their building blocks, i.e., between electrons, holes, and nuclei. Thus, understanding the many-body interaction between these fundamental building blocks holds the key to advancing fundamental science and, at the same time, directly leads to applications. Attosecond science now provides an entirely new view into the quantum many-body interaction of these microscopic building blocks.

What challenges can attoscience address differently?

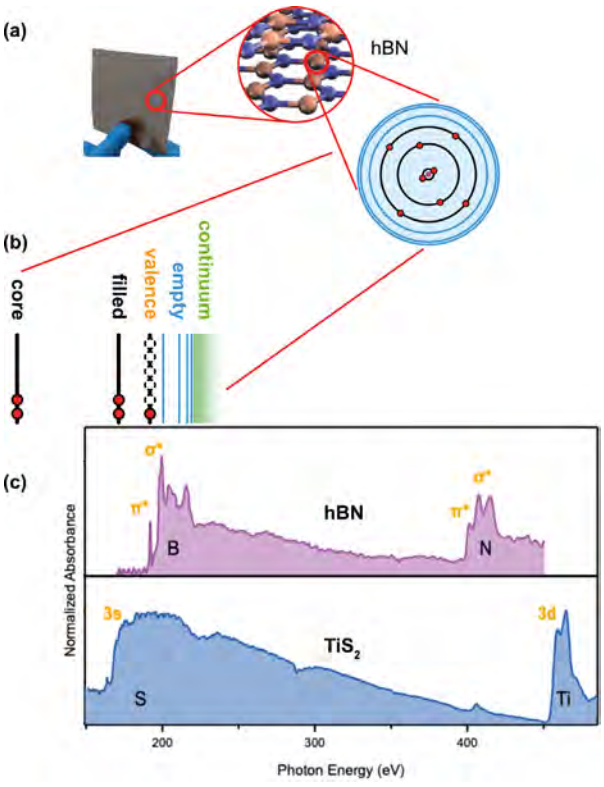
Substances and materials consist of nuclei, each with an element-specific electronic structure, whose spatial arrangement is balanced by the electrostatic force. Since the myriad of electrons and nuclei are all coupled, any change, such as electronic excitation due to absorption of light or lattice vibration due to heat, will cause a rearrangement of the electrons and nuclei. Quantum mechanics governs this rearrangement and occurs over a large temporal scale until it reaches equilibrium again. Unsurprisingly, understanding the genuine many-body interaction on a fundamental level is one of the most challenging problems of contemporary physics. Nevertheless, addressing this challenge is paramount to understanding fundamental physics phenomena, developing new substances, or improving the efficiency and functionality of modern devices. To address the challenge, scientists often simplify the problem by separating the time scales of electrons and nuclei; electrons are on the attosecond scale, and nuclei

are much slower. Thus, many perceive attoscience as a field that only aims to visualize electronic dynamics. However, the most important and open questions revolve precisely around the strong coupling on comparable and ultrafast time scales, thus depriving us of the justification to separate the time scales between electrons, holes, and nuclei. Examples of such cases are quantum phase transitions, such as superconductivity or superfluidity, or non-adiabatic dynamics and conical intersections occurring in ubiquitous processes like DNA protection or retinal vision.

So why can Attoscience address such problems?

Attosecond technology has advanced to a level where we can control electric field waveforms on the attosecond temporal scale at soft X-ray energies. So why is this a real game-changer? The short temporal duration of attosecond pulses enables time-resolved measurements on an unprecedented level on the intrinsic timescale of electronic motion, the attosecond scale. However, by focusing only on the temporal aspects, we discard the real power of attosecond pulses, namely the coherent, broad bandwidth that interrogates a material or molecule within attoseconds. So, what is the difference? Energy resolving the attosecond interaction allows us to read the electronic fingerprint of a substance in real time! What does this mean? If we know the electronic occupation and how it changes, then we can hope to ● ● ●

▼ **FIG. 1:** Coherent ultrabroad spectrum of an isolated attosecond SXR pulse [1] measures the complete electronic structure of all components of a material. Examples are shown for hBN and TiS_2 [2].



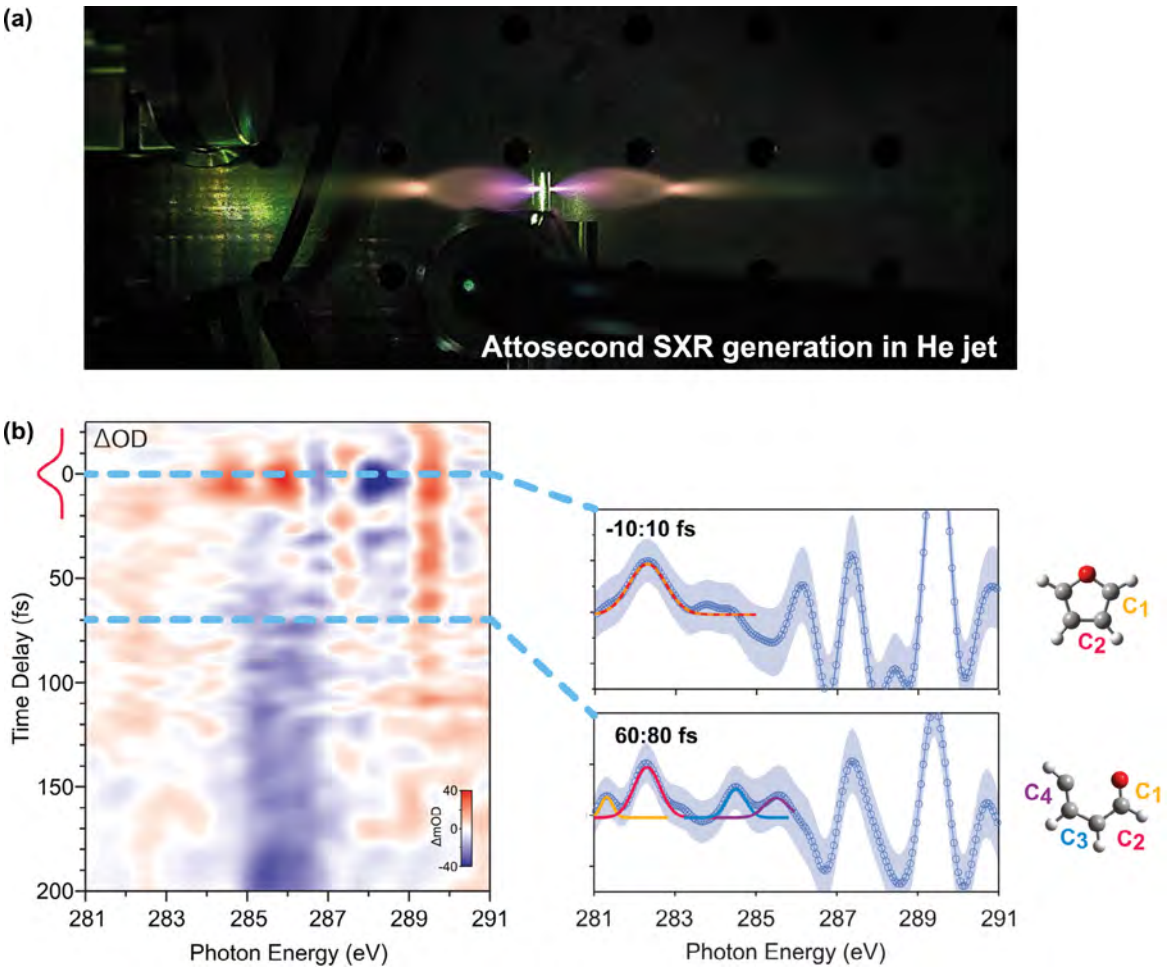
understand and alter its function. So, where does the many-body physics come in? Attosecond pulses come with a broad, coherent bandwidth. Hence, we can "see" the entire electronic structure simultaneously and with unprecedented attosecond temporal resolution. Figure 1 shows one of our measurements in which the broad coherent spectrum identifies the electronic structure of all elemental constituents of hBN and TiS_2.

Moreover, the spectrum probes all unoccupied states as well, thus providing information about the holes. Whenever nuclear motion occurs, such as molecular vibration or phonon motion, this shift, especially in the unscreened, unoccupied electronic states, is due to Coulomb interaction. Thus, the time and energy-resolved interrogation of materials provides a complete map of the coupled interaction between carriers and nuclei, *i.e.*, their many-body interaction [3-6].

This is the real power of attosecond science, and no comparable insight is available from any other single method.

Where is the information?

We now have to learn how to extract information from the time-energy measurement. Here, we do not have to start from zero since we can draw from developments in optical pulse metrology, which evolved from non-energy-resolving autocorrelation measurements to time-frequency characterization, which provides amplitude and phase information. For the same reason, attempts are underway to establish 2D spectroscopy in the XUV regime. Figure 2 shows an example of a recent measurement at ICFO in which we use pulses of 23 - 165 as duration [1] with a continuous and coherent spectrum ranging from 100 to 600 eV [4]; Fig. 2a shows a picture of the high-pressure He jet in which the attosecond soft-X-ray pulse is generated. Figure 2b shows some results from a time-energy map of a pump-probe measurement. A phase-stable pulse excites the heterocyclic ring molecule furan, and the time evolution of the system is probed at increasing time intervals with the soft-X-ray attosecond pulse to answer the question of how non-adiabatic dynamics leads to the breaking of a chemical bond and opening of the ring. The lineouts (Fig. 2b, right-top; red/yellow dashed) show the generation of a hole at ~282 eV by exciting an electron from the ground state by the optical pump pulse. After about 60 fs, the ring opens, and rovibrational signatures imprint onto the spectrum. Analysing the time evolution of such changes reveals the entire time history of carrier/nuclear dynamics.



▸ **FIG. 2: (a)** Picture of a 2-micron, sub-2-cycle laser pulse interaction in high-pressure He to generate an isolated 23-165 as pulse [1]. **(b)** The entire electron-nuclear dynamics is encoded in the time-energy measurement of furan. Lineouts show the absorption of a pump photon (top). It takes ca 60 fs for the bond to break. The 4 non-identical bonds of the open ring appear as 4 peaks in the SXR spectrum.

Outlook

We have shown the first results using attosecond time-energy measurements to address many-body problems in condensed matter and chemical physics. The attoscience field is rapidly developing, providing an entirely new angle into the many aspects of light-matter interaction at the extremes of time and energy. Questions such as the origin of a superconducting phase transition, high-temperature superconductivity, phase changes, or non-equilibrium states of matter are now in range [7]. The described methodology may yield new insight into long-standing problems related to technological issues, such as inefficient energy conversion and storage, into light-harvesting or catalysis. By using the quantum coherence of the light-matter interaction, we have tools at hand that are about to address many exciting fundamental and applied questions of tremendous relevance for science and technology. ■

Acknowledgements

The author wishes to thank his present and former students, postdocs and colleagues, in particular for the tremendous efforts when establishing our laboratory in 2007 from scratch. Further thanks go to the colleagues and staff at ICFO and ICREA for their support over the many years.

About the Author

Jens Biegert is the ICREA Professor of Attoscience and Ultrafast Optics at ICFO – The Institute of Photonic Sciences. Since 2007 at ICFO, his research focus lies on the investigation of the real-time quantum dynamics in atoms, molecules and solids. This research employs home-built cutting-edge laser technology, attosecond soft x-ray pulses and single electron diffraction. He currently serves on the Board of Directors of Optica (formerly the Optical Society, OSA), he served as Director of the EU's Laserlab-Europe Association and ARIE – The Analytical Research infrastructures of Europe. He is a Fellow of Optica, The American Physical Society, German Scholarship Association, received the Thousand Talents Award, Allen Prize, Bessel Prize of the Humboldt Foundation and holds an ERC Advanced Grant.

References

[1] S. L. Cousin *et al.*, *Phys. Rev. X* **7**, 041030 (2017).

[2] A. M. Summers et al, *Ultraf. Sci.* **3**, 4 (2023).

[3] T.P.H. Sidiropoulos *et al.*, *Phys. Rev. X.* **11**, 041060 (2021).

[4] N. Saito et al, *Optica* **6**, 1542 (2019).

[5] H. Timmers *et al.*, *Nature Comm.* **10**, 3133 (2019).

[6] T.P.H. Sidiropoulos *et al.*, *Nature Comm.* **14**, 7407 (2023).

[7] E. Rowe *et al.*, *Nature Phys.* **19**, 1821 (2023).