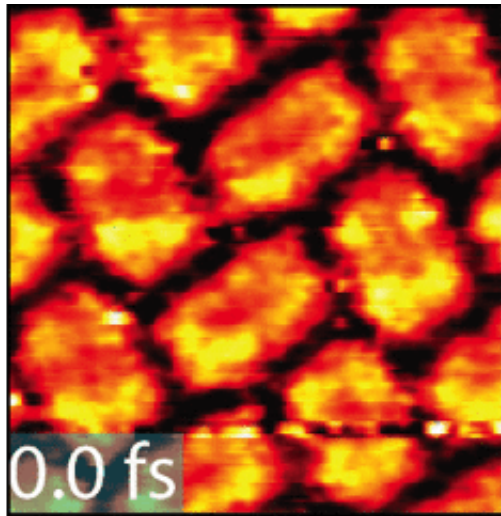


# Subfemtosecond imaging of quantum electronic coherences in molecules

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Fluctuating electrons: The film shows where the most energetic charge carriers of the organic dye PTCDA are located. The PTCDA molecules are bordered by areas of low electron density (black) at the beginning of the image. The brighter the colour, the higher the electron density: The electrons therefore move back and forth between the centre of the molecules and their edges. Credit: Manish Garg / MPI für Festkörperforschung

In order to better understand (and possibly control) fast chemical reactions, it is necessary to study the behavior of electrons as precisely as possible—in both space and time. However, up to now, microscopy methods have delivered only either spatially or temporally sharp images. By cleverly combining established techniques of tunneling microscopy and laser spectroscopy, a team led by Klaus Kern, Director at the Max

Planck Institute for Solid State Research in Stuttgart, has now overcome these obstacles. Using their atomic quantum microscope, they can make the movement of electrons in individual molecules visible.

It is essential not only for understanding [biological processes](#) (e.g., plant photosynthesis) to map the [electron dynamics](#) in [molecules](#) but also for many technical applications such as the development of solar cells or new types of electronic components. Until now, imaging methods have sometimes delivered images that are difficult to reproduce—or even contradictory. This is because they cannot map the fast electrons directly but rather must resort to techniques that can only reconstruct the behavior of the electrons.

Although modern microscopy techniques offer almost unlimited possibilities, certain compromises must be made. For example, scanning tunneling microscopy with a resolution of one tenth of a picometre ( $1 \times 10^{-12}$  m) allows extremely sharp images of individual atoms to be taken. However, this is slow and cannot capture the electron dynamics in a material. On the other hand, optical methods with ultra-fast [laser](#) pulses can detect electron movements in the attosecond ( $1 \times 10^{-18}$  of a second) range. However, they can provide only coarse, washed-out spatial images—far removed from the atomic resolution possible with scanning tunneling microscopes. The typical electron dynamics and laser pulses are in the range of a few hundred attoseconds.

### **Camera made of scanning tunneling microscope and attosecond pulses**

"For the past few years, we have been working on combining these two techniques in such a way that we can use their strengths without introducing their weaknesses," says Manish Garg, research group leader at the Max Planck Institute for Solid State Research. To do this, the researchers had to couple tried and tested scanning tunneling microscopy

with state-of-the-art laser technology. In a scanning tunneling microscope, a wafer-thin, atomically thin tip travels just above a conductive surface. Thanks to the quantum-physical tunnel effect, electrons can flow between the surface and the microscope tip—even if there is no direct contact. For example, a molecule on a surface can gradually be shaved off atom by atom.

The new microscopy technique uses laser pulses in order to modulate the tunnel current by selectively exciting the electrons in the material. "This must be done extremely quickly. Otherwise, thermal effects come into play and make the measurements impossible," explains Alberto Martin-Jimenez, who played a key role in the experiments. Unfortunately, the necessary ultra-fast laser pulses in the attosecond range are not available "out of the box." However, thanks to the rapid development of laser technology in recent years, researchers have now been able to generate precisely the right pulses. Two years ago, Garg and Kern demonstrated the function of such an atomic quantum microscope for the first time.

## **New perspective on biophysical processes, solar cells, and transistors**

They have now been able to directly observe electron movement in molecules with this one-of-a-kind instrument. With the help of the ultra-short pulses, the electrons in the molecule can be excited to jump between the different orbitals. This was noticeable in the tunnel flow. The highlight of the new technique is being able to fire two minimally time-delayed pulses in quick succession at the molecule to be investigated with an exact time interval and scan it in the process. If this procedure is repeated several times and the time interval between the pulses varied, an image series that reproduces the behavior of the electrons in this molecule with atomic accuracy is obtained. The fast [laser pulses](#) thus provide the information about the electron dynamics

whilst the scanning tunneling microscope precisely scans the molecule.

"This allowed us to directly map the dynamics of electrons in molecules—how they jump from one orbital to another—for the first time," says Garg. "This basic technology provides completely new possibilities for directly observing quantum mechanical processes such as charge transfer in individual molecules and thus better understanding them." It is still not possible to predict the possible areas of application for such a quantum [microscope](#). Especially in charge transfer processes, which play a crucial role in many biophysical reactions as well as in solar cells and transistors, it could provide crucial new insights.

**More information:** M. Garg et al, Real-space subfemtosecond imaging of quantum electronic coherences in molecules, *Nature Photonics* (2021). [DOI: 10.1038/s41566-021-00929-1](https://doi.org/10.1038/s41566-021-00929-1)

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