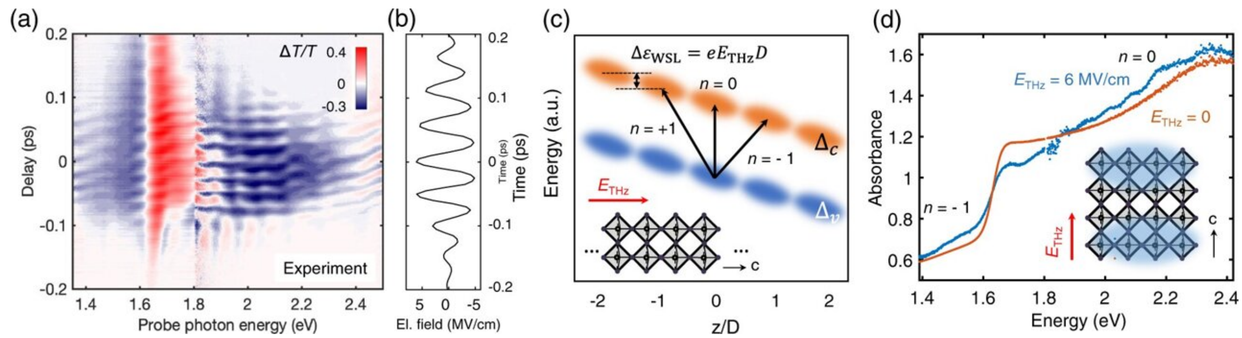
Wannier-Stark localization achieved in polycrystals

September 30 2021



Experimental observation of the transient Wannier–Stark localization and the visualized diagram. a Experimental differential transmission spectra on a polycrystalline film of MAPbI3 perovskite at room temperature, as a function of delay time of probe pulses after THz pump pulses. The THz pulses have a peak field strength of 6 MV/cm and a center frequency of 20 THz; the probe pulses have photon energy of 1.4 ~ 2.4 eV. b Temporal profile of the applied THz bias transient. c Schematic picture of Wannier–Stark localization. In the presence of strong external fields along the c axis, electronic states (orange: conduction band, blue: valence band) are localized to a few layers of ab plane and energetically separated by ΔεWSL = eETHzD between adjacent lattice sites. Black arrows depict the interband transitions within the same site (n = 0) and between different sites (n = ±1). d The absorbance with and without the external transient biasing. The Wannier–Stark localization effectively reduces the 3D electronic structure into 2D layered structure along the ab plane, as depicted in blue together with the simplified 3D structure. In case of ETHz = 6 MV/cm in considering the lattice constant D of 12.5 Å, ΔεWSL = eETHzD is estimated to be 750 meV, consistent with the spectrum showing that the absorption band of n = −1 and n = 0 are ~750 meV apart. Credit: DOI: 10.1038/s41467-021-26021-4

Scientists from Paderborn University, the Max Planck Institute for Polymer Research and the University of Konstanz have succeeded in achieving a rare quantum state. They are the first to have demonstrated Wannier-Stark localization in a polycrystalline substance. Predicted around 80 years ago, the effect has only recently been proven—in a monocrystal.

Until now, researchers assumed this localization to be possible only in such monocrystalline substances which are very complicated to produce. The new findings represent a breakthrough in the field of physics and could in future give rise to new optical modulators, for example, that can be used in information technologies based on light, among other things. The physicists have published their findings in the well-respected technical journal, *Nature Communications*.

Stronger and faster than lightning

The atoms of a crystal are arranged in a three-dimensional grid, held together by chemical bonds. These bonds can, however, be dissolved by very strong electric fields which displace atoms, even going so far as to introduce so much energy into the crystal that it is destroyed. This is what happens when lightning strikes and materials liquefy, vaporize or combust, for example. To demonstrate Wannier-Stark localization, the scientists' experiments involved setting up electric fields of several million volts per centimeter, much stronger than the fields involved in lightning strikes. During this process, the electronic system of a solid—in this case, a polycrystal—is forced far from a state of equilibrium for a very short time.

"Wannier-Stark localization involves virtually shutting down some of the chemical bonds temporarily. This state can only be maintained for less than a picosecond—one millionth of one millionth of a second—without destroying the substance. Once the electric field inside the crystal is strong enough, the chemical bonds towards the field are deactivated, rendering the crystal briefly as a system of unbonded layers. Chaos reigns. The phenomenon correlates with drastic changes to the electronic structure of the crystal, resulting in stark changes to optical characteristics, in particular, high optical nonlinearity," explains Paderborn University's Professor Torsten Meier, who was responsible for the theoretical analysis of the experiments. Nonlinear effects can give rise to new frequencies, for example, without which the targeted manipulation of light needed for modern telecommunications would not be possible.

The move from monocrystalline to polycrystalline

The effect was first demonstrated three years ago using intense terahertz radiation in a particular crystalline structure, involving the precise arrangement of the atomic structure, in a gallium arsenide crystal. "This precise arrangement was necessary for us to be able to observe field-induced localization," explains Meier, who simulated and described the experiments carried out at the University of Konstanz in 2018. Now the physicists have gone one step further.

"We wanted to investigate whether polycrystalline perovskite, commonly used in solar cells and LEDs, can also be used as an optical modulator," says Heejae Kim, team leader at the Max Planck Institute for Polymer Research. Optical modulators target the characteristics of light to make it usable in additional ways. Among other things, they are used in telecommunications, LCDs, diode lasers and materials processing. However, until now their manufacture has been not only costly, but also almost solely restricted to the field of monocrystals. Polycrystals such as perovskite could change that, being used as affordable modulators with a broad range of applications in future.

Simulations prove conjecture

"In spite of the random orientation of the individual crystallites, the small building blocks within the polycrystal, we were able to observe clear results that correspond to those characteristic of Wannier-Stark localization," continues Kim. The simulations carried out in Paderborn later confirmed these findings. Meier explains, "Although the sample is polycrystalline, it appears that the field-induced changes in the optical characteristics are dominated by a particular orientation between the crystallites and the electric field."

Over and above the first realization of Wannier-Stark localization in a polycrystalline substance, there is one thing that is particularly worthy of note: The intensity of the field required to observe the effect is considerably lower than in the monocrystalline gallium arsenide. According to Kim, "This is a result of the atomic structure of perovskite, that is, of the coincidence of a high lattice constant—the distance between the atoms—and a narrow spectrum in a particular crystal orientation. The researchers' future plans involve investigating more fully this extreme state of matter at the atomic level, researching additional substances and examining further applications of the effect.

More information: Daniel Berghoff et al, Low-field onset of Wannier-Stark localization in a polycrystalline hybrid organic inorganic perovskite, *Nature Communications* (2021). DOI: 10.1038/s41467-021-26021-4

Provided by Paderborn University