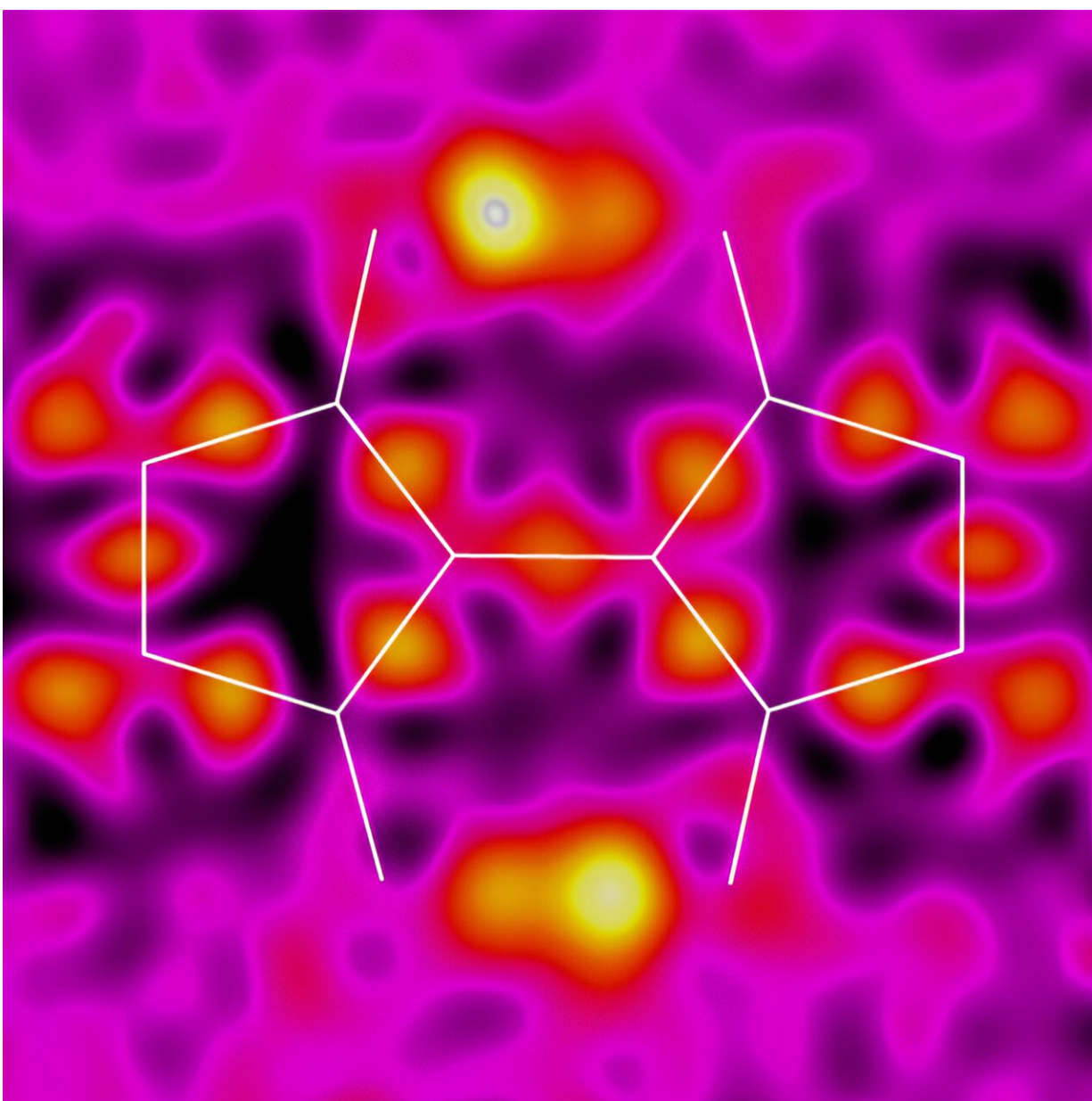


Method to make synthetic derivative of natural indigo may inspire future electronic devices

February 1 2024



Single-crystal x-ray analysis captures the two protons (bright yellow spots, top and bottom) moving between two positions on the molecule. Credit: Royal Society of Chemistry, [CC BY 3.0 DEED](#)

Chemists at RIKEN have developed a method for making synthetic derivatives of the natural dye indigo that doesn't require harsh conditions. This discovery could inspire advances in electronic devices, including light-responsive gadgets and stretchy biomedical sensors.

Semiconductors based on [organic molecules](#) are attracting much interest because—unlike conventional rigid semiconductors based on silicon—they could be flexible, ductile and lightweight, opening up new possibilities for designing [semiconductor devices](#).

Organic molecules also have the advantage of realizing a broad range of structures. "Organic semiconductors have flexibility in molecular design, enabling them to adopt new functionalities," says Keisuke Tajima of the RIKEN Center for Emergent Matter Science, who led the research [published](#) in *Chemical Science*.

To explore this potential for enhanced electronic function through molecular design, Tajima and his team investigated a molecule related to indigo, called 3,3-dihydroxy-2,2-biindan-1,1-dione (BIT). "This project started with a simple question: Can protons and electrons move in concert in the solid state?" says Tajima.

Proton-coupled electron transfer—in which the motion of electrons is linked to that of protons—is often considered critical for realizing efficient electron transfer in biological systems. If it can be incorporated

in organic [solid-state materials](#), it could lead to semiconductors with unique dynamic properties. Until now, however, no solid-state material displaying proton-coupled electron transfer has been demonstrated.

Tajima and his team have now found that BIT and its derivatives undergo unusual rearrangements in their structures involving double-proton transfer, which may lend them unique capabilities as electronic functional materials.

Tajima identified BIT and its derivatives as promising materials for solid-state proton-coupled electron transfer, because the molecule incorporates two protons that appear ideally positioned to hop from one position to another during electron transfer.

Until now, making BIT required [harsh conditions](#) that severely restricted the range of derivatives that could be made. Members of the team developed a room-temperature approach that enabled the synthesis of several BIT derivatives under much milder conditions.

With BIT derivatives in hand, the team explored the molecules' properties. "The most difficult part was to prove that the protons in BIT undergo proton transfer between molecules in the solid state," says Tajima. In collaboration with RIKEN experts in X-ray crystallography and solid-state [nuclear magnetic resonance](#) (NMR), the team demonstrated that the two [protons](#) do rapidly exchange their positions.

Calculations suggest that proton transfer is indeed coupled with [charge transport](#); the team's next target is to confirm this coupling experimentally. "We don't know if the presence of a proton will enhance charge transport, but as fundamental physics it could open interesting avenues," says Tajima.

More information: Kyohei Nakano et al, Synthesis of

3,3'-dihydroxy-2,2'-diindan-1,1'-dione derivatives for tautomeric organic semiconductors exhibiting intramolecular double proton transfer, *Chemical Science* (2023). [DOI: 10.1039/D3SC04125E](https://doi.org/10.1039/D3SC04125E)

Provided by RIKEN

Citation: Method to make synthetic derivative of natural indigo may inspire future electronic devices (2024, February 1) retrieved 4 October 2025 from <https://phys.org/news/2024-02-method-synthetic-derivative-natural-indigo.html>

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