

Novel spectroscopic technique for exploring relaxor–ferroelectric materials

June 26 2023

Polarization Behavior in Compositionally Graded Relaxor–Ferroelectric Crystals

Relaxors (lead-based B-site complex perovskites) display large dielectric and piezoelectric responses

But, understanding their inhomogeneous structures and relaxation dynamics is challenging

Polarization-angle-resolved Raman mapping for relaxor–ferroelectric materials

Ti-composition gradient $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ -ferroelectric- PbTiO_3 (PMN-xPT) solid-solution system

Raman mapping in X-Y

M_B phase

Top view

- ✓ Incorporates direction of polarized light
- ✓ Change in spontaneous polarization direction in the M_B phase
- ✓ Enhanced dielectric response at morphotropic phase boundaries

Polarization-angle-resolved Raman mapping can provide a better understanding of the relationship between the complex structure of crystals and their material properties

Polarization behavior in a compositionally graded relaxor–ferroelectric crystal visualized by angle-resolved polarized Raman mapping
 Tsukada et al. (2023) | *Communications Physics* | DOI: 10.1038/s42005-023-01219-8

Lead-based B-site complex perovskites, also referred to as "relaxors," exhibit large dielectric responses due to their complex structures. Understanding their atomic arrangements and inhomogeneous structures is, therefore, challenging. In this study, researchers from Japan used polarization-angle-resolved Raman spectroscopy on a compositionally graded crystal, identifying specific regions with enhanced charge accumulation. This information provides crucial insights into the material's behavior and provides opportunities for improving the design and performance of ultrasound devices. Credit: Yasuhiro Fujii from Ritsumeikan University, Japan

The exploitation of polarization or charge separation in ferroelectric materials has led to remarkable advances in various fields, such as the development of new ultrasound diagnostic devices. Prominently, these ferroelectric materials have led to piezoelectric devices capable of transforming electric signals into mechanical motion. Understanding how the electric polarization is arranged and fluctuates in a material is key to building better devices. However, disorders in the atomic arrangement along with their inhomogeneous structures can lead to irregular charge distribution in specific regions, posing a fundamental challenge to the development of ferroelectric materials.

To visualize the effect of disorders on polarization behavior, researchers led by Professor Yasuhiro Fujii from Ritsumeikan University, Japan, have developed an innovative polarization-angle-resolved Raman microscope. This patented technique builds upon the principles of Raman microscopy and involves directing a focused laser beam onto a sample and analyzing the [scattered light](#) to understand the molecular structure of materials. Unlike traditional microscopes, the new technique incorporates a rotating half-wave plate into the microscope setup to consider the effects of light polarization without the need to rotate the sample under study. This novel approach produces spectra with different light polarization directions at each point in the sample under study.

Combining the spectral data makes it possible to identify not only the vibration states of atoms but also the vibration directions in the material.

Now, in a study published in the journal *Communications Physics* led by Prof. Shinya Tsukada from Shimane University and Prof. Fujii, researchers have used this technique to observe the arrangement of the [electric polarization](#) and the time scale of its fluctuation in a piezoelectric lead magnesium niobate $[\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3]$ -lead titanate $[\text{PbTiO}_3]$ or PMN-PT crystal, which is used in diagnostic ultrasound equipment, revealing the reason for the large dielectric constant.

"The development of this polarization-angle-resolved Raman microscope along with the advancements in analytical techniques can enable the incorporation of polarization information into the existing Raman imaging data and allow a deeper understanding of material properties," explains Prof. Fujii, speaking of the rationale behind the development of this technique.

One notable characteristic of PMN-PT crystals is their pronounced dielectric and piezoelectric response at the boundaries that separate different phases in the material. The specific composition of the PMN-PT crystal, particularly the concentration of titanium (Ti), can affect the formation and characteristics of phase boundaries. To investigate the effect of Ti mixing ratios on the dielectric properties, the researchers imaged a $62.7 \times 15.0 \times 0.3$ PMN-PT crystal sample with the newly developed setup for Raman mapping in the microscope.

The Ti content varied from 27.0 mol% to 38.0 mol% along the length of the sample, giving rise to three [distinct phases](#): a monoclinic (type B) phase where the Ti content ranged from 27 mol% and 29.2 mol%, a monoclinic (type C) phase where it went up to 34.5 mol%, and a tetragonal phase with a high Ti content of 34.8–38.0 mol%.

On analyzing the Raman spectra corresponding to different light polarization values at each point in the sample, the researchers observed abrupt changes in the intensity of the Raman peaks only for the monoclinic type B phase. Moreover, they also noted a distinct change in the direction of spontaneous polarization in this phase. The spectra revealed a slower relaxation (reorientation of the electric dipoles in response to a thermal perturbation) of the material's polarization closer to the phase boundary between the monoclinic (type B) and (type C) phases. This, in turn, indicated that the realignment of the dipoles occurs at a reduced rate, enabling the material to store a large amount of charge and display enhanced dielectric response at this phase boundary.

"We found that the ability of the relaxor–ferroelectric material to store a significant amount of electric charge is due to the slow response of nanometer-scale electric polarization to the external voltage," highlights Prof. Fujii.

In summary, the observation of this characteristic property of relaxor materials highlights the capability of the polarization-angle-resolved microscope to provide polarization information, which could help optimize a material's dielectric performance. In particular, the insights into the polarization behavior of PMT-PT could enhance the development of relaxor materials with improved ultrasound detection and generation properties for next-generation diagnostics.

More information: Shinya Tsukada et al, Polarization behavior in a compositionally graded relaxor–ferroelectric crystal visualized by angle-resolved polarized Raman mapping, *Communications Physics* (2023).
[DOI: 10.1038/s42005-023-01219-8](https://doi.org/10.1038/s42005-023-01219-8)

Provided by Ritsumeikan University

Citation: Novel spectroscopic technique for exploring relaxor–ferroelectric materials (2023, June 26) retrieved 2 May 2024 from <https://phys.org/news/2023-06-spectroscopic-technique-exploring-relaxorferroelectric-materials.html>

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