1-1-2. Transient Absorption Spectroscopy of Solid Materials

Overview

Transient absorption spectroscopy of solid materials monitors absorption changes using a time-controlled probe after photoexcitation, enabling analysis of non-equilibrium processes from femtoseconds to milliseconds. It reveals exciton and carrier dynamics, lattice responses, and defect states, and is widely applied to organic crystals, semiconductors, and perovskite crystals, providing fundamental insights for optical materials and device development.

Current Status and Frontiers

Transient absorption spectroscopy of solid materials is widely used as a powerful technique for directly observing photoexcitation processes and carrier dynamics in materials. Historically, it originated from the development of flash photolysis by George Porter and colleagues (awarded the 1967 Nobel Prize in Chemistry) and was further advanced by Ahmed Zewail's establishment of femtosecond pump–probe spectroscopy, leading to the 1999 Nobel Prize in Chemistry. As illustrated in Fig. 1, an excitation pulse and a probe pulse are irradiated onto a sample with a controlled time delay, and changes in transmitted light intensity due to new absorption transitions are measured as a function of delay time. Transient absorbance is evaluated according to the Lambert–Beer law using $\Delta OD = \log(I_0/I)$. In principle, the time resolution is determined by the pulse width rather than the detector response, meaning that pulse compression directly improves time resolution.

Traditionally, transient absorption spectroscopy was conducted using transmission optics for optically transparent samples, but with the increasing diversity of solid materials, techniques utilizing specular reflection and diffuse reflectance[1] are gaining importance. For opaque materials, polycrystalline samples, powders, and thin films,

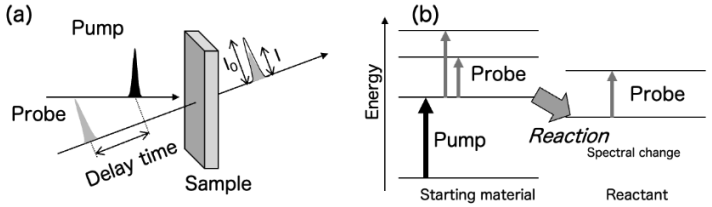

Figure 1. Principle of transient absorption spectroscopy.

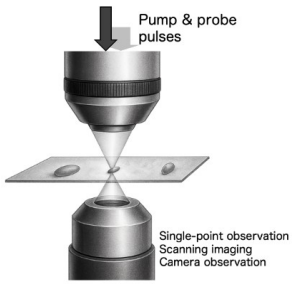

Figure 2. Diagram of a transient absorption microscope.

diffuse reflectance methods are effective for non-destructive, high-sensitivity detection of transient surface and interfacial responses, providing information unattainable through transmission measurements. Driven by growing demand for nanoscale structures and microdevice research, transient absorption spectroscopy combined with microscopy optics has rapidly advanced[2,3] (Fig. 2). By integrating femtosecond lasers with confocal microscopy or scanning probe techniques, it is now possible to visualize localized absorption responses on submicron scales, enabling investigations of local electron and lattice dynamics in microcrystals, single nanowires, and two-dimensional materials.

Furthermore, transient absorption spectroscopy in the extreme ultraviolet to X-ray range using free-electron lasers[4] and high-harmonic generation sources is attracting attention as a novel means of observing photoinduced chemical bond and atomic structural changes in real time. These technological innovations are being applied to a wide range of materials, including semiconductors, organic thin films, perovskite solar cells, and quantum materials, advancing understanding of carrier relaxation, defect dynamics, and photoinduced phase transitions. In addition, with the growing scale of experimental datasets, machine learning and statistical analysis are increasingly introduced, accelerating the extraction of new physics from complex spectroscopic responses and their application to materials design.[5]

References

1. A. Furube, Z.S. Wang, K. Sunahara, K. Hara, R. Katoh, M. Tachiya, *J. Am. Chem. Soc.*, **132**, 6614–6615 (2010).
2. N. Gross, et al., *J. Phys. Chem. C*, **127**, 14557–14586 (2023).
3. T. Katayama, A. Yamamoto, Y. Fujita, Y. Akagi, P. Koinkar, A. Furube, *Jpn. J. Appl. Phys.*, **62** SG1029 (2023).
4. M. Harmand, M. Cammarata, M. Chollet, et al., *Sci. Rep.*, **13**, 18203 (2023).
5. T. Gutberlet, H.T. Chang, S. Zayko, M. Sivis, C. Ropers, *Opt. Express*, **31**, 39757–39764 (2023).

Future Perspectives and Directions

With further advancements in spatial and temporal resolution as well as AI-driven analysis, it is expected that the understanding of photoexcitation processes at the atomic scale will deepen, accelerating the design and creation of novel photo-functional materials.

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