

Optical switching of magnetism

Researchers realized a magnet that can optically switch the polarization plane of light by 90 degrees. *Nature Photonics* asked Shin-ichi Ohkoshi how his group achieved this feat.

■ What was the motivation for your work?

The ferromagnetic properties of materials that contain the well-known blue pigment Prussian blue were discovered 50 years ago. Since then, their magnetic and optical properties have attracted great interest. These materials exhibit a huge variety of crystal structures and absorption spectra, which vary sensitively according to the incorporated metal ions. Despite their name, they are not always blue — they can be other colours, such as red and yellow. Inspired by these characteristics, we investigated the dependence of the magnetic and optical properties of Prussian blue materials on their crystal structure. We expected this to spawn unprecedented magneto-optical phenomena — one of the aims of our laboratory. As you know, the Kerr effect is one such phenomenon; it is applied to readout from magneto-optical recording materials. However, the Kerr effect in such materials typically rotates the plane of polarization of light by less than one degree, and hence a complex optical system is required for precise detection. To overcome this problem, we have been working on developing a magneto-optical material that exhibits a large magneto-optical effect at visible wavelengths.

■ What were the keys to achieving a polarization rotation angle of 90 degrees?

There were two key points. First, photoinduced phase I has a tetragonal crystal structure that has a high symmetry. Second, the nonlinear susceptibility for second-harmonic generation $\chi^{(2)}$ in photoinduced phase I is zero because the transition is forbidden at the excitation wavelength. If the tensor elements of $\chi^{(2)}$ were non-zero, the nonlinear susceptibility for second-harmonic generation would compete with that for magnetization-induced second-harmonic generation, $\chi^{(2)\text{mag}}$, and the polarization rotation angle would be less than 90 degrees. We honestly didn't expect to realize such a gigantic rotation angle, because no method had been developed to make the value of $\chi^{(2)}$ zero. So, we planned to design materials whose values of $\chi^{(2)\text{mag}}$ were enormous compared with $\chi^{(2)}$. If we could achieve this, we estimated that the rotation angle would be at most about 20 degrees. Fortunately, the present material satisfied the two key



Asuka Namai, Kenta Imoto, Shin-ichi Ohkoshi, Hiroko Tokoro and Marie Yoshikiyo (left to right) experimentally demonstrated spin-crossover-induced second-harmonic generation, light-reversible spin-crossover ferromagnetism and photoswitching of magnetization-induced second-harmonic generation. They also elucidated the mechanisms behind these effects.

points, allowing us to realize 90-degree polarization switching.

■ What was the most difficult aspect of this work?

Determining the crystal structures of photoinduced phases I and II, which were produced by laser irradiation, was challenging. In the first place, there was no guarantee that the sample consisted of only one photoinduced phase; it might have been made up of multiple phases. We repeated the X-ray diffraction measurements several times after laser irradiation, and thoroughly investigated which diffraction peaks are sensitive to the photoinduced phase transition. We found some diffraction peaks whose intensities were correlated with the laser beam intensity. Assuming that the sample consists of only one photoinduced phase, we analysed the change in the X-ray diffraction spectrum and determined the crystal structures before and after each phase transition. To obtain the magnetic space group of the sample, we had to determine both its crystal structure and its space group. Of course, we could infer the crystal structure to a certain extent from that of the low-temperature phase. However, such analysis of the physical properties of the sample is speculative. To remove this ambiguity, we had to be 100% certain of the crystal structure after the photoinduced phase transition.

■ What applications is this magneto-optical material expected to have?

Potentially, the magneto-optical material could be applied to high-speed optical switching, as it can switch the polarization of light by 90 degrees on optical stimulation. In this study, we demonstrated this ability at low temperature. If it could be demonstrated at room temperature, we could realize a transient photoinduced phase transition. We expect the speed of the transient transition to be on the order of a nanosecond. Furthermore, the light remains linearly polarized, which is not possible by other techniques. For example, the polarization plane of the light can be rotated by 90 degrees by propagating linearly polarized light in a transparent glass tube and inducing the Faraday effect by applying a magnetic field, but magnetic resonance in magnetic materials causes linearly polarized light to become elliptically polarized.

Another possible application is optical memory. In this magneto-optical material, the switching angle can be controlled between 0 and 90 degrees by varying the photoinduced magnetization. By using intermediate angles, n -ary systems ($n = 4, 8, 16, \dots$) will be possible, in addition to binary systems ($n = 2$); this will enable new multinary notation high-density magneto-optical memories. Currently, the photoinduced phase transition occurs at a temperature of around 20 K. It is challenging to develop magneto-optical materials that operate at room temperature.

■ What future plans do you have for this research?

The material in this study has a rather complicated molecular structure, but we have demonstrated the essence of the photoinduced magnetic phase transition and have found out how to rotate the polarization of light by 90 degrees. We are now assessing whether the same principles can be applied to other kinds of materials, such as oxides. We hope to design and realize novel optical functional materials based on ubiquitous materials.

INTERVIEW BY NORIAKI HORIUCHI

Shin-ichi Ohkoshi and co-workers have an Article on the optical switching of magnetism on page 65 of this issue.