# Report for MERIT Long-Term Overseas Dispatch

Department of Advanced Materials Science

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**Place**: Laboratory for Multifunctional Ferroic Materials, D-MATL, ETHZurich

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**Theme**: Observation of spin-driven ferroaxial order via second harmonic generation

**Overview**

I stayed with Prof. Fiebig's group at ETH Zurich, Switzerland, and performed experiments about the observation of ferroaxial order originating from spin orderings using second harmonic generation (SHG).

**Research**

Ferroaxial order is a new class of ferroic order characterized by rotational distortions in crystals, which has the same symmetry as the electric toroidal dipole corresponding to the vortex arrangement of electric dipoles. This ferroaxial order has so far been studied as a structural order, caused by the displacement of ions in a crystal. More generally, however, it is defined by mirror symmetry breakings and does not necessarily require direct ionic displacement. In the present study, therefore, we expected ferroaxial order to be manifested by ordering of spins in crystals, and attempted to observe it.

The material under investigation is $NdB_4$, which has a tetragonal crystal structure belonging to the space group $P4/mbm$ and exhibits non-colinear antiferromagnetic (AFM) ordering below the transition temperature $T_0$ = 17.2 K. The magnetic point group in the AFM phase is $2'/m$. The mirror symmetries preserved in the paramagnetic phase are broken, and ferroaxial order is expected to emerge. At the same time, ferrotoroidal order originating from the breaking of spatial and time-reversal symmetries also appears. In this study, we tried to observe these symmetry breaking-derived ordered states using SHG.

First, the temperature dependence of the SHG intensity was measured. Finite signals were observed below $T_0$, and the signal intensity behaves like an order parameter with respect to temperature. Then, the spatial distributions of SHG were measured below $T_0$, and a characteristic pattern of dark lines was observed. This is presumably caused by the interference of SHG tensor components of opposite signs at the domain boundaries. Measurements of the azimuth angle dependence of the SHG intensity between the neighboring domains did not show the mirror symmetry breaking expected in the ferroaxial order. Therefore, the observed domain structure is considered to correspond to ferrotoroidal order. A possible reason for the absence of ferroaxial order is that the main order in $NdB_4$ is ferrotoroidal order, and SHG derived from ferroaxial order may have been masked by that derived from ferrotoroidal order. On the other hand, the ferrotoroidal domain structure was clearly visualized.

$NdB_4$ is a highly conductive metal, and observations of antiferromagnetic and ferrotoroidal domains in such metallic compounds are very limited. We are currently considering publishing the results obtained in this study about the visualization of ferrotoroidal domains in the antiferromagnetic metal.

**Life on the campus**

The laboratory of the Prof. Fiebig group located on the ETH Hönggerberg campus on a hill some distance away from the center of Zurich. The laboratory building has a large lawn behind it, from which a magnificent view of Switzerland could be seen (Fig. 1).

Fig. 1. ETH Hönggerberg Campus.

Prof. Fiebig group was relatively large, with nearly 10 doctoral students alone, and weekly seminars in the group were attended by more than 20 members. The research topics are diverse, including thin-film synthesis by PLD, imaging using nonlinear optics, time-resolved measurements using terahertz waves, etc. The door of Prof. Fiebig's room was often closed in the mornings to allow him to concentrate on writing, but in the afternoons he gladly opened the door for us to come in. In addition, a fully automatic espresso machine was installed in the group's common space, so we could drink delicious coffee at any time. There were many opportunities to discuss and chat over coffee, which played an important role in enriching life in the group.

Although this stay was relatively long (5 months), due to the current difficult situation, the use of liquid helium, which is essential for my experiments, was not possible during the first two months. I remember that I was suddenly informed of that one week before the start of my stay and travelled there in a state of anxiety. Even so, I was able to spend some meaningful time taking part in time-resolved measurements, which I had not initially planned to do, and constructing the necessary optical system from scratch. The stay was not without its troubles, as the entire city suffered a power failure at the very moment when I finally became able to start measurements and was about to transfer liquid helium into a cryostat. Anyway, it was the first opportunity for me to perform nonlinear optical measurements and I was able to learn the basics over time.

**Life in Zurich**

As my stay in Zurich was during the winter, I had to endure the bitter cold. Zurich is a very safe city and I was able to walk outside even late at night without feeling unsafe. The high cost of living goes without saying, so I cooked for myself almost every day and brought my own lunch box after making a lot of food the night before. As the Christmas season was included in my stay, I was able to have some unique European experiences, such as visiting a Christmas market on the weekend and attending a Christmas party in the lab. group. Authentic cheese fondue had a strong wine flavor.

**Acknowledgements**

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