

Report on MERIT Overseas Dispatch Program

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1. Overview

From January 3, 2016 to March 31, 2016, I stayed and studied at Prof. Toomas Rõõm, and Prof. Urmas Nagel's group (THzLab) in National Institute of Chemical Physics and Biophysics, Tallinn, Estonia.

2. Research content

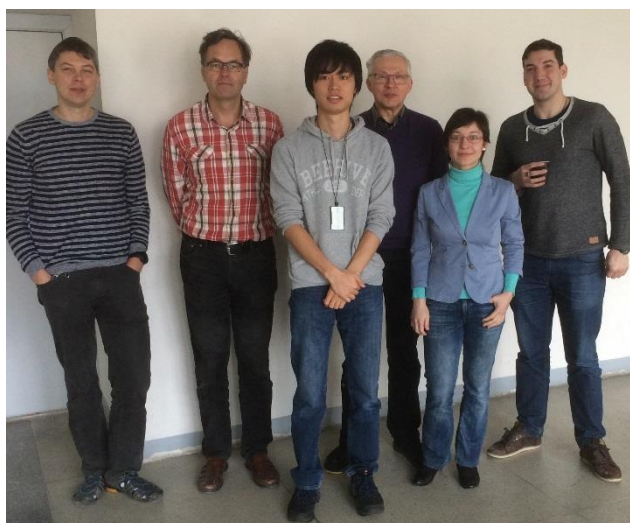
Recently, it has been found that magnetic materials show ferroelectricity universally and they have been studied actively from the perspective of the control of magnetization (ferroelectric polarization) by electric fields (magnetic fields). It is known that in multiferroics, ferroelectricity originates from spin order via mechanism such as spin-orbit interaction, etc. Therefore, there is strong correlation between their magnetism and ferroelectricity. As a phenomenon showing this correlation, a elementary excitation of spin called electromagnon is well known, which is driven by electric field components of lights. The resonant frequency is mainly at terahertz frequency region. Multiferroics have taken much attention because it may be possible to realize ultrafast control of magnetization or ferroelectric polarization by exciting electromagnons which are unique modes in multiferroics. THzlab is one of the world leading group which study various multiferroics from the spectroscopic perspective using their equipment which can measure high-resolution optical spectra in microwave ~ far-infrared region (0.06 ~ 8 THz) with applying the magnetic field up to 17 T

In this dispatch, I purposed learning the terahertz spectroscopy in THzlab and study electromagnons in ferrimagnetic compounds, $\text{BaFe}_{12}\text{O}_{19}$. The main equipment consist of a spectrometer and a cryostat with a superconducting magnet which can apply magnetic fields up to 17 T. Samples and the magnet are cooled by filling the cryostat with liquid helium. Magnetic fields can be applied in both Faraday and Voigt configuration. The measurement system is based on a Martin-Puplett interferometer, a mercury arc lamp as a light source and a Si bolometer cooled down to 300 mK as a detector. Spectra of lights transmitting a sample can be obtained by Fourier transformation of measured interferograms. In the beginning of staying, the measurement system had problem, which the signal to noise ratio was too bad. Then, I helped improving it for the first month. In particular, we restored bad connections of wires and took a preamplifier and a digitizer etc. into metal boxes to prevent noise due to external electromagnetic waves. After improving the signal to noise ratio, I measured the spectra for ferrimagnetic $\text{BaFe}_{12}\text{O}_{19}$. At

room temperature, $\text{BaFe}_{12}\text{O}_{19}$ has a hexagonal crystal structure with a point group of $6/mmm$. Fe^{3+} ions occupy 6-oriented octahedral, 4-oriented tetrahedral or 5-oriented bipyramid sites. Below 723 K, Fe^{3+} ions in the high-spin state ($S = 5/2$) are ordered, resulting in the ferrimagnetism at room temperature. Also, Fe^{3+} ions in 5-oriented bipyramid sites are displaced from the center position toward the positive or negative direction of the c axis, which induce local electric dipoles along the c axis. The bipyramid FeO_5 constitute a triangular lattice in the c plane with antiferroelectric dipole-dipole interactions, resulting in frustration in the lattice. Consequently, $\text{BaFe}_{12}\text{O}_{19}$ shows a quantum paraelectric behavior without long-range electric ordering down to extremely low temperature. Therefore, $\text{BaFe}_{12}\text{O}_{19}$ is a unique compound, which is both ferrimagnetic and quantum paraelectric. If there are any interactions between the local ferroelectric polarization and the magnetization, it is possible that a phase transition, such as formation of a long-range order of ferroelectric polarization, gets occurred by breaking the balance between local electric-dipole interactions and frustration in the lattice by applying magnetic fields at low temperature. If gets occurred, we may observe any excitations of electromagnons etc. Then, I measured the transmittance spectra of $\text{BaFe}_{12}\text{O}_{19}$ at 4 K with applying magnetic fields in Faraday and Voigt configuration. However, no absorption peaks except an usual magnetic excitation were observed. The first possibility of the reason is that interactions between the ferroelectric polarization and the magnetization are small, so that any phase transitions or any changes didn't occurred at the magnetic field applied in the experiments. Another one is that the measurement temperature (4 K) was much high. In fact, in the previous study [S.-P. Shen *et al.*, *Nat. Commun.* **7**, 10569 (2016).], the temperature dependence of the specific heat showed anomaly around \sim several hundred mK.

3. Living at Tallinn

Tallinn, the capital of Estonia, which is one of the Baltic states, is a small city with population of about 400 thousand. The national language is Estonian, but almost Estonian people can speak English, so I had few linguistic problems at daily life. The eating habits are mainly based on sausage and meat. Although I felt nervous whether the food suited my taste before the dispatch, most food was really delicious. In the holidays, I went sightseeing to many spots such as the old



With people of Lab.

town of Tallinn which is a world heritage, Riga in Latvia, Vilnius in Lithuania, Helsinki in Finland, Stockholm in Sweden, etc. by bus or ship. I concerned about the staying because it was the first time for me to live by myself at a foreign country for a few months, but I had few troubles and enjoyed staying. However, sometimes I felt troubles in communication because of my poor English, so I keenly felt the importance of improving my English.

4. Acknowledgement

I really appreciate Prof. Rõm, and Prof. Nagel and THzLab's people for letting me stay at THzlab and giving me a great experience. Also, I would like to thank to Associate Prof. Kida for contacting to Prof. Rõm. Finally, I appreciate MERIT, Prof. Okamoto, Prof. Arima for supporting the dispatch.