Review of the PhD thesis “Magnetoelectric multiferroics: From static via dynamic magnetoelectric effect to nonlinear light-matter interaction by Jakub Vít

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The thesis of Jakob Vit covers spectroscopic investigations of numerous magnetoelectric materials with the focus on optical directional anisotropy. This topic has recived a lot of interest recently due to the novel possibilities for both fundamental physics and applications. In general the work is adequate and well presented. The specific comments on individual chapters are presented below.

1 Theoretical background of magnetoelectric multiferroics

The introductory chapter first gives the definitions of multiferroics and magnetoelectrics and also points out the symmetry relations and basic constraints between different components of magentoelectric and conventional susceptibilities. It continues by classification of multiferroics within the thesis and mentions a possible manifestation of different multipole orders in magnetoelectric susceptibilities. A good overview of known magnetoelectric coupling mechanisms is given then. The last part presents the connection between the time-reversal symmetry of magnetoelectric tensor and the optical directional anisotropy, which is the main experimental observable of the thesis. A practical magnetoelectric sume rule is also derived. The chapter concludes with the general symmetry conditions to observe optical directional anisotropy and its classification.

2 Experimental methods and techniques

This chapter gives a brief yet adequate description of the principal experimental methods used in the thesis. Some photographs or diagrams would be a worthy addition to aid in the descriptions of the different apparatus. Equation (2.2) is missing a square.

3 Isothermal control of THz nonreciprocal directional dichroism by electric field and magnetic-field tilt in the multiferroic $Ba_2CoGe_2O_7$

The chapter gives a very clear example of the engineered realization of the electric field control of the magnetic state in a multiferroic. It starts with a list of general prerequisites to achieve this goal and consistently demonstrates their fulfillment in a particular multiferroic $Ba_2CoGe_2O_7$ in specially chosen geometries. Different sets of well planned experiments demonstrate the manipulation of magnetic domains by either electric field or tilting of magnetic field or both of them. Very thorough analysis of the experimental results is augmented by the microscopic spin model. The final result of this analysis is the comparison of different scenarios of the domain behavior. Most of the observed effects can be explained within reasonable assumptions and the remaining discrepancies are well documented.

4 Electromagnons in hexaferrites: spectroscopy and microscopic selection rules based on the exchange-striction mechanism

This chapter covers the characterisation of serval hexaferrite compounds using static (magnetisation/polarization) and dynamic (THz/IR/Raman) techniques. The measurements are comprehensive, particularly the investigation of the Y-type compound. The results and discussion is convincing making the work a good contribution to the study of electromagnets in these systems.

In particular a number of electromagnon excitations are identified and characterised as a function of temperature. It would be a nice addition to see plots of the peak position, spectral weight and damping as a function of temperature so that the profiles can be easily observed and compared with the static measurements and emergence of the different magnetic phases.

This would be particularly interesting for the double peak structure of the electromagnon in Y-hex. It seems that under field the two peaks have a slightly different dependence. Is this true? Could the two peaks be coming from different modulations of the magnetic structure i.e. a mix of TC and ALC or TC with an extra longer range modulation on top?

The frustrated magnetic structure is mentioned a few times. Is this geometric or exchange driven frustration? A diagram showing the frustrated geometry would be nice to see.

Could the shift in the polar phonon mode observed in the ceramic z-hex sample be due to strain? If the small crystallites of the ceramic are under strain this might explain the small observed change in electric polarisation compared to the single crystal data from the literature.

5 Non-linear THz absorption by electromagnons in multiferroic hexaferrites

This chapter investigates the evolution of the electromagnon excitation in the z-hex and y-hex samples under the application of intense THz pulses from two separate free electron laser sources. With the recent advancement of such accelerator based sources there is a lot of potential for new and interesting non-linear measurements at THz frequencies. At the same time however, with new techniques comes new challenges and much work still needs to be done on the best practises when using these new sources. What are the best systems to look at and how should the data be analysed?

While the results of this chapter are inconclusive, important work is done on considering how to interpret the data and what can be done to improve measurements in the future. It therefore is a solid contribution to this new emerging field.

As is pointed out towards the end of the chapter, pump-probe methods may give more sensitive results. This usually involves observing the polarisation state of a transmitted or reflected optical laser pulse from the sample. I wonder what the optical properties of the samples are under to assess if such measurements are feasible. Would the sample transmit optical frequencies for example?

6 Selection of magnetoelectric domains by static and oscillating electromagnetic fields in $LiCoPO_4$

The chapter intoduces the idea of magnetoelectric domain switching by intense oscillating electromagnetic fields of an optical beam. The idea is tested in lithium-cobalt phosphate system which can be polled by static electric and magnetic fields. The domain population can be checked by either optical directional anisotropy at magnetoelectric resonances or by static magnetoelectric effect.

Although the reliable poling by free electron laser beam has been demonstrated, the effect turned to be more complicated than initially expected. The improved experimental scheme and the number of comprehensive experiments involving polarization dependence upon poling static fields, poling optical fields and the probe optical beam has shown that only the same type of domain is always selected. A thorough analysis of obtained results has lead to the conclusion that the extrinsic effects are responsible for the observed THz poling. A model involving inversion-breaking temperature gradient and the time-reversal breaking propagation of magnetoelectric excitations is put forward to qualitatively explain the observed poling polarization dependence. Despite the rather speculative character of the model, it is consistent with all observed symmetries of the THz poling.

Overall, the presented work has substantially contributed to the new topic of domain control by intense radiation even though no clear understanding of mechanisms behind this effect is achieved yet.

7 Appendix

Some nice further details regarding the analysis of the high field data amongst other things.

Conclusion of the reviewer

Overall, the ideas presented in the thesis are ambitious and relevant in the field. The desired effects were generally observed, although the physics behind them often turned out to be more complicated than originally expected. Nevertheless the analysis was always extensive and well documented. Overall, the work is a solid contirbution to the research field and is definitely suitable for the PhD defense.