

I thank Prof. Kamarás for the review. In her review, there are several assessments, suggestions or questions. I will try to address all of them:

- This intention to include everything, however, makes the thesis too long and somewhat uneven. It seems that the intention to document every experiment won over the requirement to put them in a consistent frame and extend the introduction beyond the one included in typical journal publications. It would have been preferable to concentrate on finished and consistent work, and giving less space to speculations about experiments that did not show the expected outcome or where the results cannot be explained at the moment. This would have made the thesis much more readable.

I agree that the thesis could have been much more readable and consistent. My intention was to include also some results which would not be otherwise published because our level of understanding is not sufficient to publish these results in a scientific journal. This necessarily created a space to speculations and made the thesis less readable and consistent.

- ...a clear line has to be cut between what has been in fact achieved and what has to be done in the future.

I think it is clear what was achieved in the thesis and I included several suggestions what to do in the future, which are clearly written in the future tense. I would have to see a specific example(s) to deal with this criticism.

- Chapter 5 seems a little speculative, searching for possible nonlinearity, and not simply asking the question " what happens at high intensities?" . In other words, the model is first and the search is for signs in the experiment that support the model.

I completely agree that Chapter 5 is speculative, and all flow of it may seem weird to a reader. This was caused by historical reasons – our understandings and interpretations changed during experiments and writing, and since a lot of time was invested, I decided not to rewrite the chapter from the beginning. I admit that if I was about to write the chapter now, I would have written it in a different way.

- In Chapter 6, the ultimate experiment foreseen, using intense THz fields for poling, did not give the expected result. The reasons were suspected to lie in the experimental setup, most probably thermal effects connected to the sample holder, and rather preliminary explanations are put forward. As the thesis contains enough clear and well-resolved results, these could have been left for later.

Based on this statement, I realized that my message about the analysis of the light poling was possibly not fully conveyed. I wrote Chapter 6 in a chronological and honest way, mentioning what was originally expected and that results were different. This probably made the story too complicated. For the future of science, it is completely irrelevant what the "ultimate" experiment foreseen was, but it is important what the actual results were, while I think that the obtained results are significant.

Let me summarize the main result and stress its significance: the heat flow, coming from the temperature gradient, causes the effect of poling. The temperature gradient was caused by the experimental setup and asymmetric contact to the sample holder, but this does not deteriorate the result itself. It just means that the temperature gradient is necessary for the effect, thus the crystal is exposed to some spatially varying field. Please note that temperature gradient (or other inhomogeneity such as strain gradient) is necessary also for some intrinsic effects like Seebeck or Peltier effects, which provide information on fundamental material properties.

The effect I observed is probably connected to the magnetoelectric anisotropy in diffusive transport [Phys. Rev. Lett. **94**, 016601, (2005)] (Ref. [82] in the thesis), which I also mentioned in the thesis. As

the observed effect is a hallmark of a novel phenomenon, I consider it as one of the most significant result of the thesis. I admit that the results reported in the thesis are preliminary. Nevertheless, I give clear ideas what to measure in the future (and I have already measured that), which proves my understanding of the topic. Since I spent an enormous time by light poling of  $\text{LiCoPO}_4$ , I think it was highly desirable to include this research in the thesis. However, I understand that Chapter 6 was too long for a reader. I decided to write the chapter in such a chronological and honest way to show progress in my knowledge and my ability to solve problems.

- What is missing and should have been added more emphasis instead, are the contributions to the experimental arrangements only sporadically mentioned in various chapters; the experimental innovations should have been summarized in Chapter 2.

My contributions to experimental arrangements were rather ordinary – I did not produce any state-of-art innovations. Therefore, I did not consider as important to put more emphasis on them. Nevertheless, I agree that experimental innovations should have been summarized in Chapter 2. Therefore, I give the summary of my contributions to the experimental arrangements here.

Firstly, I assembled a time-domain THz spectroscopy setup based on Teraflash spectrometer [J. Infrared Millim. Terahertz Waves **35**, 823–832 (2014)] (Ref. 143 in the thesis), allowing to exchange receiver and emitter antennas, whereby allowing to measure the directional dichroism as it is defined. The setup is displayed in Figure 1.

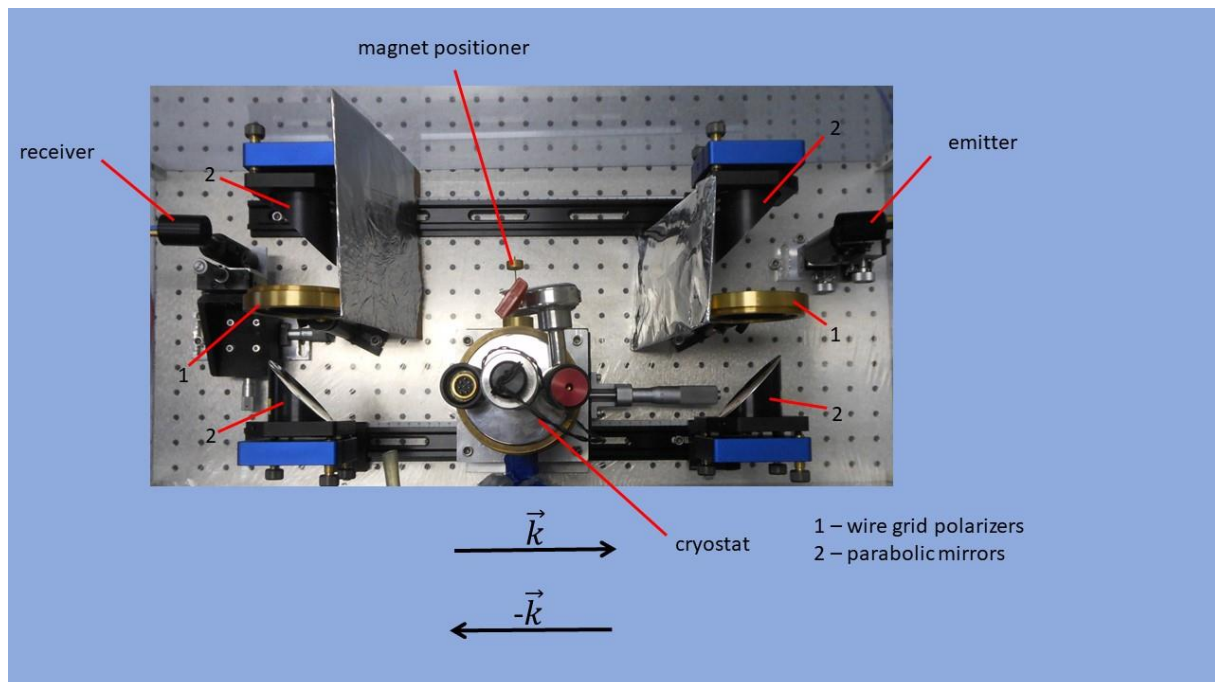


Figure 1: THz spectroscopy setup I aligned at Budapest University of Technology and Economics, allowing to exchange the emitter and the receiver, whereby allowing to measure the directional dichroism as it is defined: The light can propagate in one ( $\vec{k}$ ) or opposite ( $-\vec{k}$ ) direction. Different components of the setup are marked.

Secondly, I assembled the setup for measuring quasi-static magnetoelectric effect by the modulation technique, depicted in Figure 2. This setup is designed in the way to allow illuminating the sample from both sides in the direction perpendicular to both applied electric and magnetic field.

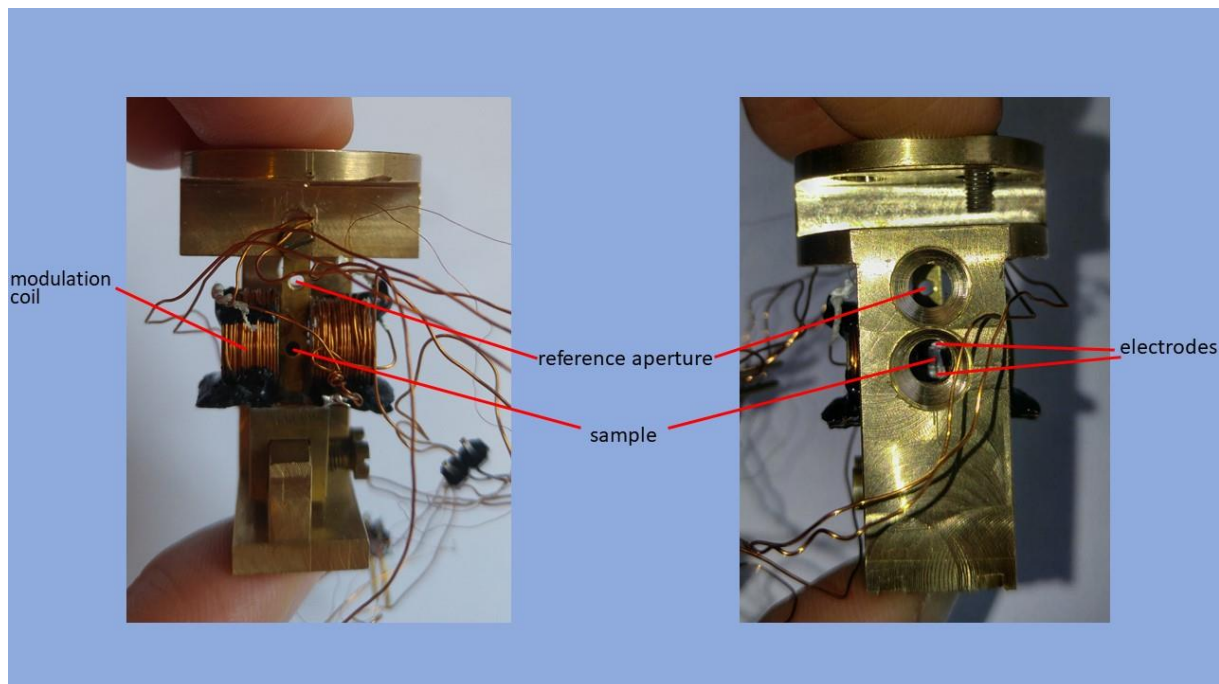


Figure 2: The setup for measuring quasi-static magnetoelectric effect by the modulation technique, photographed from opposite sides. Different components of the setup are marked.

- As to the broader context, the principal mechanism that connects all phenomena is domain dynamics and the change of domain population induced by poling effects. However, no detailed introduction on this very important and fundamental process is given in Chapter 1, except references to publications.

I agree that the change of domain population induced by poling effects is a mechanism occurring in several chapters. I did not include anything about this topic, but here let me briefly summarize the basics: When energy of some domain becomes smaller than energy of another domain, the second domain is transferred into the first one. Since the domains are typically separated by an energy barrier (coming from the magnetic anisotropy), the global minimum is not achieved and meta-stable domains may persist. If I included these few general sentences in Chapter 1, it would not bring any insight, so I decided to discuss this within the chapters related to results, in which specific problems are discussed.

The domain dynamics is definitely an important and broad topic for discussion. Nevertheless, it is beyond the scope of this thesis. For example, in the case of electric- and magnetic-field control of domains in  $\text{Ba}_2\text{CoGe}_2\text{O}_7$  (Chapter 3), I assume that only the static energetic balance is important, and domain dynamics is very fast, thus, irrelevant for the observed effect. In contrast, domain dynamics is possibly relevant in case of poling  $\text{LiCoPO}_4$  by the electromagnetic radiation. Nevertheless, my understanding of the poling process is limited. The pattern formation at the critical point is a difficult, active field of research. Note that the poling occurs in the vicinity of the Néel temperature where domains are strongly fluctuating and the order parameter is small, so experimental observation of the poling process would be probably impossible. In conclusion, the known low-temperature antiferromagnetic domain dynamics is not relevant for the topics of the thesis, so I did not include it in Chapter 1. Note that I did not use any technique of domain visualization and the performed experiments cannot be directly related to domain dynamics.

- The term ” magnetoelectric annealing” is incorrectly used throughout the thesis. Annealing is a process where a sample is heated, kept at some higher temperature and then let it cool slowly. The term is applied correctly when describing preparation details (page 31: ” ... the samples for static measurements were annealed in an oxygen atmosphere at 900 °C for 7

days” , page 65: ” hexaferrites are usually annealed in oxygen atmosphere” ). In case of magnetoelectric materials with the Néel temperature above room temperature, e.g. Cr<sub>2</sub>O<sub>3</sub>, the use of ” annealing” is justified, but with these low-TN materials, where the thermal treatment means simply the cooling of the sample, it is not (e.g. page 56: ” ME annealing from high temperatures” ). In the publications, this expression is not present, ” cooling below TN” is written instead. This would have been more fortunate to use throughout the dissertation and in Thesis point 4 as well.

Firstly, there are two terms – “annealing” and “magnetoelectric annealing”, which are completely distinct. The term “magnetoelectric annealing” is used in a topical review of Fiebig from 2005 [*J. Phys. D: Appl. Phys.* **38** R123 (2005)], which has over 4000 citations and is considered as one of the fundamental papers of the field. Therefore, “magnetoelectric annealing” can be considered as an established term in the field of magnetoelectrics and multiferroics. Its meaning is a synonym of the “magnetoelectric poling”, which means cooling down through the phase transition in electric and magnetic fields. Although it is distinct from the conventional annealing, the two processes have common features: in both cases the temperature is increased and decreased to reach reorder the domains or the crystalline defects.

What I see as my mistake is that I did not explain the term “magnetoelectric annealing” in the thesis more in detail, and I used this term as a synonym of “magnetoelectric poling”, while I cited literature in which the term “magnetoelectric annealing” was not used – this can cause a confusion to a reader. I also used both expressions, while I should have introduced the two terms as synonyms in the beginning and then use only one of the expressions.

- While I highly appreciate the comparison with the theoretical background being evaluated, I would not call the discussion of electromagnon strength in Section 4.3.5 ” quantitative” . It is at most semi-quantitative, explaining trends in the intensities of certain modes with magnetic field.

I completely agree with this critic.

- In some of the figures, the use of colors is not optimal, the curves cannot be easily assigned to the legend (Fig. 1.4, Fig. 3.5), the ” small black arrow” in Fig. 6.9 is really tiny, inconsistencies occur between the figure captions and the text (Fig. 4.3).

In case of Fig. 1.4, a reader may not infer that arrow colors correspond to the terms in equations. I thought it is intuitive, but probably I should have added an explicit description into the figure caption. In case of Fig. 3.5, I agree that the curves should have been thicker. I also agree that the small black arrow in Fig. 6.9 should have been bigger.

I suspect that there may be several inconsistencies between figure captions and the text. However, I cannot comment the case of Fig. 4.3 without knowing what exactly is inconsistent.

- The References section, unfortunately, was not compiled with enough care. Many items lack bibliographic details: items 39, 43, 90, 140, 147, 221, 233 and 252 are incomplete (this can seem small compared to the 252 items overall, but they include even papers by the closest collaborators), the others are also often inconsistent (upper-lowercase, journal names used either abbreviated or full, issue number given or not, date as month or year only). I suggest to add a corrected bibliography that will be deposited together with the thesis, in standardized form (minimum content: standard abbreviation of journal name, volume, page or article number, year). Automatic export from journal databases is not sufficient in this case.

I agree with this critic. I asked a representative of Budapest University of Technology and Economics whether the corrected bibliography can be deposited together with the thesis. If yes, I will correct it.

- p. 35. Raman spectroscopy: what is meant by "the effect is off-resonant"? Is it possible to apply Raman spectroscopy under resonant conditions in this type of measurements?

Here, I simply meant that the incoming radiation is not resonant with the studied excitation. I admit that such formulation is inaccurate, since it may evoke well-established expressions like "off-resonant Raman scattering" and "resonant Raman scattering". The latter means that the laser is in resonance with an electronic excitation, while studied excitations have always (for both off-resonant and resonant Raman scattering) lower energy than the photons coming from a laser – this is indeed what I wanted to say.