Chapter 4

Conclusions and Outlook

In this work, we have settled the theory for Second Harmonic Generation from antiferromagnetic interfaces. The importance of the antiferromagnetic oxide interfaces for the modern technology and material science has been outlined in the Introduction. The main conclusion of this work is that SHG possesses unique capabilities to address antiferromagnetic interfaces. In particular, the symmetry analysis of the nonlinear magneto-optical response, described in the Chapter 2 of this work, demonstrates the potential of SHG to:

- distinguish magnetic phases of the surface, in particular detect surface antiferromagnetism,
- distinguish detailed spin configurations of the AF surface,
- indicate for the distortions of the lattice
- image the AF surface domains

As proposed by us in the Subsec. 2.3.8, these results can be experimentally obtained by the appropriate choice of geometry. The analysis was applied both to stand-alone monolayers and to double layers (which represent fully the semi-infinite material) and is thus complete.

In addition to the symmetry analysis of the nonlinear magneto-optics we have, for the first time, discussed the validity of applying time-reversal symmetry to classify the SHG response. Our results show that the presence of dissipation in the frequency space makes the process of SHG dynamical and thus irreversible. Only the combination of SHG and antiferromagnetism fully reveals the complications that time-reversal operation imposes in nonlinear magneto-optics.

In the Chapter 3 of this work we have presented an electronic calculation which supports the above mentioned conclusions. The quantitative result of this calculation is that we expect the effect from the antiferromagnetic surfaces to be of the same order of magnitude as SHG from ferromagnetic surfaces, which had been proven measurable. In an experiment, one can exploit a spectral line specific for antiferromagnetism, visible even on top of a ferromagnetic background.

An important outcome of our electronic calculation is the first ever presentation of nonlinear magneto-optical spectra of NiO (Sec. 3.3). Using them, spectral lines favorable for the experimental nonlinear optics and magneto-optics can be determined. Although the used approximations do not allow for quantitative predictions of the peak heights in the spectrum, peak positions are *reliable*.

The results of the dynamics calculations (Sec. 3.4), simulating a time-resolved pump-and-probe SHG experiment, allow to make some interesting assertions about the investigated systems. In antiferromagnetic oxides, the negligible dispersion of the gap states (which are the most important ones contributing to the surface SHG) prohibits the decay of the tensor elements, unlike for metallic systems. On the other hand, the fast limit of the dynamics in our system is demonstrated to lie within a few femtoseconds. These characteristics make the antiferromagnetic oxides ideal materials for modern applications like quantum computing and magneto-optical storage, even more since no spin injection is needed.

In conclusion, our work has demonstrated that antiferromagnetic oxides, in particular NiO, have many interesting properties which will certainly draw even more attention to this class of materials, and that SHG is a quite unique tool to address these properties.

Our theory, though it yields important and reliable results, is not yet complete. The improvement of our analytic tools will go in the direction of obtaining more quantitative results. For that purpose, a more realistic estimate of transition matrix elements will be indispensable. Actually, a proper *calculation* of these transition matrix elements will make our theory fully *ab initio*, but this will not be possible in the near future. An extension to the current state of our theory which will be easier to implement but which is also quite interesting will be taking into account the three- and four hole states. Their application has already been prepared, since we have determined these states and their energies for the spherically-symmetric environment. Another point which we intend to implement is taking into account the band structure. The dispersion of the gap states, although very small, can help us in determining the long-time limit of the coherence times.

These improvements will provide an even more complete description of nonlinear magneto-optics from antiferromagnetic surfaces of nickel oxide.