Theoretical Description of X-ray Experiments on Magnetic Materials out of Equilibrium

H. Ebert, J. Braun, A. Marmodoro, and S. Mankovskyy

Department of Chemistry, Ludwig-Maximilians-University, Munich, Germany,
he@gaia.cup.uni-muenchen.de

Representing the electronic structure of an investigated material in terms of its electronic Green function is a well established and flexible starting point to calculate its X-ray absorption and optical spectra [1]. With very few exceptions, this scheme was used so far only to deal with materials in their equilibrium ground state. We present extensions of this scheme in two directions by dealing with a steady-state out-of-equilibrium situation as well as explicitly time dependent pump-probe experiments.

In the first part of the contribution we consider the case of XAS/XMCD investigations in multilayer systems subjected to a finite static electric field [2]. Our computational framework is based on the spin-polarized relativistic multiple scattering or SPR-KKR (Korringa Kohn Rostoker) Green function scheme. This approach has been extended by various authors to the steady-state out-of-equilibrium situation described by a corresponding energy dependent non-equilibrium Green function (NEGF) [3]. The NEGF has been used so far primarily to deal with transport properties. Here we demonstrate its use for the investigation of spectroscopic properties by calculating the electric field induced changes in the XAS- and XMCD-spectra of the Pd $L_{2,3}$-edges in a Co/Pd bilayer system. A corresponding extension of the XMCD sum rules allows a comparison of field induced moments as seen by XAS-spectroscopy and calculated directly on the basis of the NEGF. An extension of the scheme to deal with the spin-pumping experiments of Van der Laan and coworkers [4] is briefly discussed.

Recent developments in time-dependent density functional theory (TD-DFT) paved the way towards investigating and quantitatively interpreting, on an ab initio level, the ultrafast demagnetization processes in ferromagnetic systems caused by a strong laser pulse [5]. These time-dependent phenomena can be monitored by time-resolved spectroscopic techniques such as for example angle-resolved photo emission (ARPES) or magnetic circular X-ray dichroism (XMCD), with the latter having the additional advantage of being element-specific. A theoretical description for a time-dependent ARPES experiment has been recently worked out and implemented by the authors on the basis of the one-step model of photo emission [6]. The corresponding time evolution of the investigated system due to a pump pulse is described by means of the two-time Keldysh NEGF that is evaluated by means of the SPR-KKR Green function method. In this contribution a corresponding description for pump-probe XMCD experiments will be presented that makes use of the same theoretical framework. In addition, when compared to the previous ARPES investigations [6], the time dependence of the single-particle potentials is taken care by means of self-consistent TD-DFT calculations [5]. It should be stressed that the Keldysh NEGF formalism allows in addition to account for the impact of various relaxation mechanisms represented by a corresponding complex and energy-dependent self-energy. First numerical results for XMCD spectra at the $L_{2,3}$-edge of ferromagnetic Co exposed to a strong pump laser pulse that are based on a time dependent potential and using so far a quasi-static standard description of the XMCD experiment [1] will be presented.

References

[1] - H. Ebert, *Rep. Prog. Phys.*, **59**, 1665 (1996).
[2] - R. Kukreja et al., *Phys. Rev. Letters* **115**, 096601 (2015).
[3] - S. Achilles et al., *Phys. Rev. B* **88**, 125411 (2013); M. Ogura, H. Akai, *J. Phys. Soc. Japan* **85**, 104715 (2016).
[4] - G. van der Laan, *Physics* **8**, 83 (2015).
[5] - K. Krieger *et al.*, *J. Chem. Theory Comput.* **11** 4870 (2015).
[6] - J. Braun, R. Rausch, M. Potthoff, and H. Ebert, *Phys. Rev. B*, **94**, 125128 (2016).