Theoretical description of ultrafast laser-induced demagnetization monitored by photo-emission using the non-equilibrium Green function formalism

Hubert Ebert[1] and Jürgen Braun[1]

[1]*Department Chemie, Ludwig-Maximilians-Universität München, 81377 München, Germany*

Most theoretical schemes to describe ultrafast laser-induced demagnetization of solids are working on a model level using input parameters that might be provided by electronic structure calculations. Only recently [1] an approach based on time-dependent density functional theory (TD-DFT) was presented that allows to calculate the time evolution of the magnetisation due to a laser pulse in a parameter-free way. The applicability of the approach was demonstrated for the elemental ferromagnets but also for Heusler alloys [2]. As an alternative we suggest to use the Keldysh non-equilibrium Green function formalism, that offers a number of advantages when dealing with the dynamics of an electron system under the influence of an external perturbation. A corresponding formal basis has been worked out by us that allows in particular to describe the time evolution of the electronic system of a magnetic solid under the influence of a laser-field of arbitrary shape and intensity [3] in terms of the so-called double-time dependent lesser Green function $G^{<}(\mathbf{r},\mathbf{r}',t,t')$. As has been shown, $G^{<}(\mathbf{r},\mathbf{r}',t,t')$ can be calculated on the basis of a Dyson equation from the retarded Green function of the unperturbed system that in turn is obtained in a very flexible and efficient way using the multiple scattering or KKR formalism. The previous TD-DFT based work demonstrated that spin-orbit coupling plays a central role for ultrafast demagnetisation. To account for this, our fully relativistic approach is based on the Dirac equation. Using a combination of local spin-density approximation (LSDA) and dynamical mean field theory (DMFT) as a basis for the underlying electronic structure calculations allows an adequate treatment of correlation effects in narrow band systems that are represented by a corresponding complex and energy-dependent self-energy. The present approach also allows to include a double time-dependent self-energy within the calculations. This in principle allows to account for dynamical correlation effects but also for damping or relaxation effects due to electron-phonon coupling or other mechanisms. An interesting feature of our approach is that it can be combined in a direct way with the calculation of spectroscopic properties as required when dealing with pump-probe experiments [4]. This is demonstrated by results from a study on two-photon photo emission from ferromagnetic Fe (100) [5].

[1] K. Krieger, J. K. Dewhurst, P. Elliott, S. Sharma, E. K. U. Gross, J. Chem. Theo. Comp., **11**, 4870 (2015).
[2] P. Elliott, T. Müller, J. K. Dewhurst, S. Sharma, and E. K. U. Gross, Sci. Rep., **6**, 38911 (2016).
[3] J. Braun, R. Rausch, M. Potthoff , J. Minar and H. Ebert, Phys. Rev. B **91**, 035119 (2016).
[4] J. Braun, R. Rausch, M. Potthoff and H. Ebert, Phys. Rev. B **94**, 125128 (2016).
[5] J. Braun and H. Ebert, Phys. Rev. B **98**, 245142 (2018).