Quantum atomistic model for the ultrafast evolution of magnetic systems

O. Morandi[1], P.-A. Hervieux[1], G. Manfredi[1]

[1] Institut de Physique et Chimie des Matériaux de Strasbourg and Labex NIE,

Université de Strasbourg, CNRS UMR 7504 BP 43, F-67034 Strasbourg Cedex 2, France.

omar.morandi@ipcms.unistra.fr

The study of the out-of-equilibrium properties of strongly excited magnetic systems is today a very active area of research. The understanding of the physical mechanisms that are at the basis of the spin transfer and of the magnetic quenching in clusters of interacting molecules or solids constitutes an important challenge both from the experimental and the theoretical point of view [1]. The elementary mechanisms that lead to the modification of the magnetic order triggered by laser pulses are at the center of an intense debate. Various processes have been proposed, as for example the Elliot-Yafet mechanism for itinerant charges, the modification of the exchange splitting energy or the ultrafast diffusion of spin polarized current. The analysis of the optical response of a magnetic system could be greatly facilitate by the development of accurate microscopic models and numerical methods that are able to reproduce the complex many-body dynamics of a growing number of interacting atoms. Moreover, X-ray scanning techniques provide two-dimensional images of the magnetic distribution in a solid with the resolution of few nanometers. This opens the intriguing possibility to compare the nanometric evolution of the spin polarization detected by optical methods with the microscopic atomic spin configuration obtained by ab initio methods.

For this purpose, various theoretical and numerical approaches have been developed. A general and powerful technique is the so called “atomistic approach”, where the atom-atom interaction is modeled by the short range Heisenberg interaction and the evolution equation of every single atom is numerically solved [2]. The static and dynamic properties of systems whose size spans from few nanoparticles to a large number of atoms (bulk materials) can be reproduced. Such atomistic models are based on the classical description of the atomic spin and angular momentum provided by of the phenomenological Landau–Lifshitz–Gilbert equation. The validity of such classical description of the electron spin applied to magnetic nanosystems may be questionable. Moreover, the atomistic models are characterized by a large number of parameters that are usually obtained by fitting some knows experimental results.

In the present contribution, we propose a new atomistic-like approach where a system constituted by interacting atoms is described in a quantum framework. A cluster of atoms or, more generally, a nanomaterial is described in terms of a network of quantum spin particles interacting via exchange interaction. Moreover, the crystal band structure and exchange constants are obtained by ab initio (DFT) calculations. The evolution equations are solved numerically by applying a Monte Carlo technique.

The model takes into account the dynamical atomic correlations and the evolution of the exchange energy splitting. It is able to describe fully coherent phenomena such as the ultrafast exchange of spin of angular momentum among the atoms inside a solid and the

dissipative effects induced by interaction with the phonon bath.

In this contribution, we focus on the evolution of the total angular momentum of a nanometric material in the presence of the strong field produced by a femtosecond laser pulse. In particular, we have reproduced the ultrafast evolution of the spin and angular momentum and the transfer of magnetization between transition metal (Co) and rare earth (Tb) in anti-ferromagnetic phase after the excitation by a femtosecond laser pulse. The evolution of the spin polarization of the system is described in terms of the following processes: *(i)* the nearest neighbour atomic spin transfer driven by the Heisenberg interaction, *(ii)* the heat transfer from the kinetic degree of freedom exited by the laser to the spin network induced by the scattering of itinerant electrons *(iii)* the modification of the spin polarization induced by the spin-orbit coupling and finally *(iv)* the quenching of the angular momentum induced by the electrostatic crystal field.

One of the major results of our simulation is reproduced in Figure 1 where we depict the evolution of the total angular momentum of the two atomic species that constitute the alloy. Our simulations are in quantitative agreement with the observations made by N. Bergerard et al. [3] and shed light on the microscopic spin exchange mechanisms that are at the origin of the macroscopic evolution of the total magnetization. In particular, we discuss the difference on the dynamical evolution of the magnetization for ferromagnetic and anti-ferromagnetic alloys.

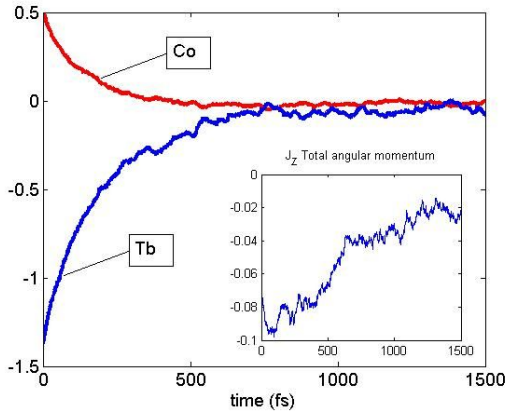

Figure 1: Ultrafast magnetization transfer between Cobalt and Terbium in anti-ferromagnetic $Co_{0.74}Tb_{0.26}$ after the excitation by a femtosecond laser pulse. The inset shows the evolution of the total magnetic momentum of the crystal.

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