



# ***Optical Characterization of Ferromagnetic and Multiferroic Thin-Film Heterostructures***

**Xin Ma**

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Advisor: Gunter Luepke, Professor of Applied Science

## **Abstract**

This thesis presents optical characterization of the static and dynamic magnetic interactions in ferromagnetic and multiferroic heterostructures with time-resolved and interface-specific optical techniques. The focus of the thesis is on elucidating the underlying physics of key physical parameters and novel approaches, crucial to the performance of magnetic recording and spintronic devices. First, time-resolved magneto-optical Kerr effect (TRMOKE) is applied to investigate the spin dynamics in L10 ordered FePt thin films, where perpendicular magnetic anisotropy  $K_u$  and intrinsic Gilbert damping  $\alpha_0$  are determined. Furthermore, the quadratic dependence of  $K_u$  and  $\alpha_0$  on spin-orbit coupling strength  $\xi$  is demonstrated, where  $\xi$  is continuously controlled through chemical substitution of Pt with Pd element. In addition, a linear correlation between  $\alpha_0$  and electron scattering rate  $1/T_e$  is experimentally observed through modulating the anti-site disorder  $c$  in the L10 ordered structure. The results elucidate the basic physics of magnetic anisotropy and Gilbert damping, and facilitate the design and fabrication of new magnetic alloys with large perpendicular magnetic anisotropy and tailored damping properties. Second, ultrafast excitation of coherent spin precession is demonstrated in Fe/CoO heterostructures and  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$  thin films using TRMOKE technique. In the Fe/CoO thin films, instant non-thermal ferromagnet (FM)  $\leftrightarrow$  antiferromagnet (AFM) exchange torque on Fe magnetization through ultrafast photo-excited charge transfer possesses in the CoO layer is experimentally demonstrated at room temperature. The efficiency of spin precession excitation is significantly higher and the recovery is notably faster than the demagnetization procedure. In the  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$  thin films, pronounced spin precessions are observed in a geometry with negligible canting of the magnetization, indicating that the transient exchange field is generated by the emergent AFM interactions due to charge transfer and modification of the kinetic energy of  $e_g$  electrons under optical excitation. The results will help promoting the development of novel device concepts for ultrafast spin manipulation. Last, the interfacial spin state of the multiferroic heterostructure  $\text{PbZr}_{0.52}\text{Ti}_{0.48}\text{O}_3/\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$  and its dependence on ferroelectric polarization is investigated with interface specific magnetization induced second harmonic generation (MSHG). The spin alignment of Mn ions in the first unit cell layer at the heterointerface can be tuned from FM to AFM exchange coupled, while the bulk magnetization remains unchanged as probed with MOKE. The discovery provides new insights into the basic physics of interfacial magneto-electric (ME) coupling.