Project Information

Project title: Magneto-optical Kerr effect of antiferromagnetic materials in external magnetic fields

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Executive summary:

We investigate the relationship between light and magnetically ordered materials, in particular, metallic antiferromagnets. Their weak response to applied external magnetic fields and THz frequency spin dynamics are interesting characteristics which make antiferromagnetic materials potential candidates for memory devices. In this context, the magneto-optical Kerr effect (MOKE) is a useful phenomenon which provides information about magnetization through optical response, however, antiferromagnetic materials with a compensated spin configuration have no net magnetization. Based on the magnetic susceptibility, the spin response under applied fields can be predicted and related to tilted spin configurations that arise in external magnetic fields. MOKE signals are, hence, present for tilted spin configurations. In this project, first-principles calculations based on density functional theory are carried out to study MOKE and magnetic susceptibility of antiferromagnetic materials. This will be useful guidance for experimentalists, e.g. to identify the wave length of maximum MOKE response in new materials.

Description of research activities and results:

Key Challenges:

Due to the weak response under applied external fields, it is difficult to control the internal spin configuration of antiferromagnets. Spin compensation renders detecting the spin information of antiferromagnetic materials using optical measurements challenging. Measurements of optical response under external magnetic fields have been developed, however, it is still necessary to understand the fundamental behavior of antiferromagnetic materials better. In order to understand basic principles experiment can benefit from theoretical guidance, e.g. to maximize the detected signal, in order to measure the response.

Why it matters:

Antiferromagnetic materials have interesting properties that can potentially lead to fast switching in future memory devices. Unlike ferromagnetic materials, they show a weak response to external magnetic fields and do not show a magnetic field outside of the material due to spin compensation. Understanding how to control internal spin degrees of freedom will be critical for applications of these materials and also for understanding them better on a fundamental level.

In this work, we use first-principles calculations to study magneto-optical properties of various antiferromagnetic metals. MOKE signals measured in experiment are usually small, on the nanoradian scale, which is easily overwhelmed by noise. Since it is difficult to tune the laser system every time to find the optimal wave length for excellent signal-to-noise ratios, first-principles calculations based on density functional theory can provide this information. Our computed spectra give useful guidance to experiment, in order to find the wave length that maximizes MOKE signals.

The results of magnetic susceptibility and Néel temperature illustrate that this first-principles computational approach can provide reasonable predictions for magnetic properties of antiferromagnetic materials. Thus, our calculations are helpful guidance to understand the behavior of antiferromagnetic materials and to develop advanced magnetic devices.

Why Blue Waters:

Even though first-principles calculations are a great opportunity to thoroughly investigate antiferromagnetic materials, optical and magnetic calculations still require significant computational resources. Unlike non-relativistic case, a fully relativistic band structure and its dielectric function calculation with magnetism are demanding processes. Spin tilted calculations are challenging because non-collinear calculations need more computational resources than collinear calculations. In addition, these require more computational resource because magnetic unit cells of antiferromagnets are usually larger than chemical unit cells. Magnon dispersion calculations for Néel temperatures also require dense Brillouin zone sampling to achieve convergence to an appropriate accuracy. In order to address those challenges, Blue Waters is well-suited due to fast communication and large amount of memory per node. Thus, Blue Waters provides a unique chance to unveil the unknown properties of antiferromagnetic materials.

Accomplishments:

In this project, wave-length dependent MOKE spectra are computed for MnPt and Fe2As. We find that MnPt shows a maximum Kerr rotation signal at 730 nm and Kerr ellipticity at 540 nm. For Fe2As case, Kerr rotation and ellipticity are maximized at 620 nm and 520 nm, respectively. At the same time, magnetic susceptibility is also computed, which can provide the spin response under applied external magnetic fields. For MnPt along c-axis which is perpendicular to the spin direction, computed magnetic susceptibility is χ=4.4 ×〖10〗^(-4) which is close to χ=4.742×〖10〗^(-4), as determined experimentally.

Based on our computational results, magnetic moments of spin-tilted magnetic-structure calculations can be converted to a response to external magnetic fields. Thus, MOKE signals under the applied field can be predicted. In order to confirm the spin configuration stability at room temperature, Néel temperature calculations are performed. Through the mean-field approximation, computed Néel temperature of MnPt is 928.6 K, while experimental measurement of MnPt finds 953 K.

List of publications associated with this work:

[1] K. Kang, A. Schleife: Optical and magneto-optical properties of antiferromagnetic metals; in preparation
[2] Kexin Yang, Kisung Kang, Zhu Diao, Arun Ramanathan, Daniel P. Shoemaker, Andre Schleife, David G. Cahill: Time-Resolved Quadratic Magneto Optical Kerr Effect Measurement on Metallic Antiferromagnet Fe2As; in preparation

Several poster and conference presentations.

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Plan for next year:

We plan to wrap up our ongoing JLESC collaboration with Edoardo di Napoli at Juelich, that is aimed at exploring a novel iterative diagonalization library, ChASE, for computation of optical properties within our Bethe-Salpeter code. We have done early testing on Blue Waters and we plan on expanding these tests, possibly towards GPU nodes.

Regarding magneto-optical properties, we are currently expanding the set of materials studied computationally, to better compare to results from our experimental collaborators, David Cahill and Daniel Shoemaker. We are planning to compute relaxed lattice structures, electronic and optical properties, magnon dispersions, and Neel temperatures. From these calculations we plan to develop a comprehensive picture of magneto-optical properties of antiferromagnetic metals. Materials to be studied include MnPt, Mn_2Au, Fe_2As, Mn_2As, Cr_2As, and Mn_3B_4. For these calculations we will use several tens of XE nodes. They have total memory requirements between several Gigabyte up to a few hundred of Gigabyte (via MPI). We expect to transfer small amounts of data in and hundreds of Gigabyte out of the Blue Waters machine. Input and output requirements are modest for all these calculations.

Resources requested: We request a total of 245k node hours and the default storage allocation in the project directory as well as the Nearline storage.

Estimated use: Q1: 20 %, Q2: 20 %, Q3: 30 %, Q4: 30 %

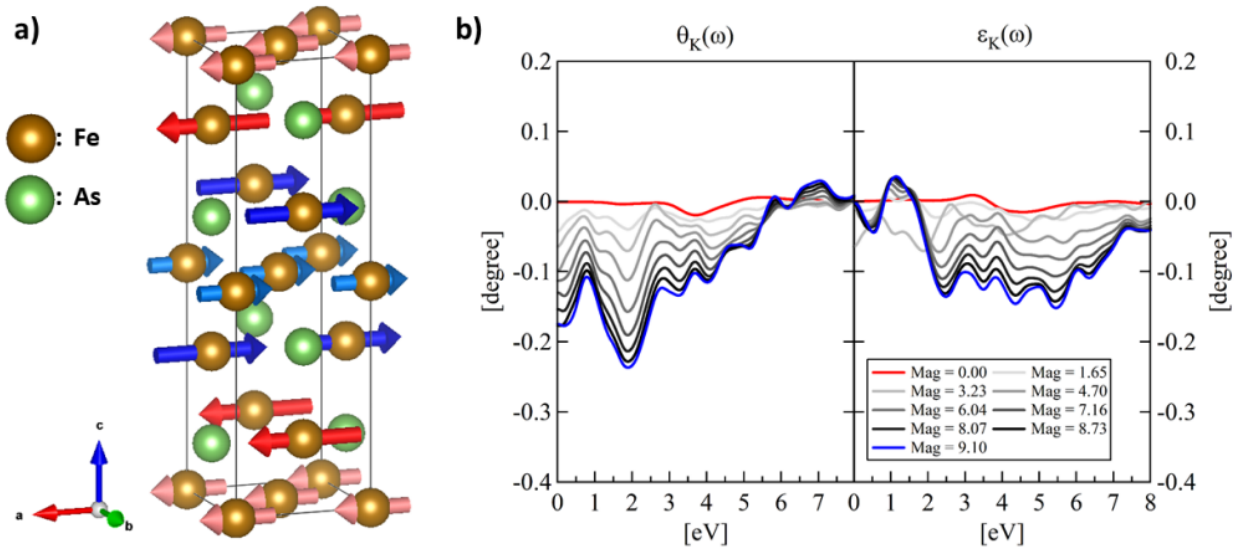

Fig. 1: a) Magnetic unit cell of Fe2As and b) Wave-length dependence of MOKE signals under external magnetic fields. Kerr rotation (θK) and Kerr ellipticity (εK) are maximized at 2 eV (620 nm) and 2.4 eV (520 nm), respectively.