Coupling of heat and spin currents at the nanoscale in cuprates and metallic multilayers

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A. Thermal transport by magnons and magnon-phonon coupling in cuprates.

Heat conduction in materials is typically mediated by thermal excitations of atomic vibrations (i.e., phonons) or thermal excitations of the electronic degrees of freedom (i.e., electrons and holes in metals and heavily doped semiconductors). However, any thermal excitation of the solid can, in principle, contribute significantly to the thermal conductivity if the heat capacity of the excitations is significant, the excitations have a large dispersion so that the group velocity is large, and the lifetime of the excitation is not too short. These conditions are met by the spin degrees of freedom in low-dimensional quantum magnets based on copper oxides (Sr14Cu24O41, La2CuO4, CaCu2O3). These materials have a seemingly unique large magnon thermal conductivity: near room temperature, the magnon thermal conductivities are comparable to the electronic thermal conductivities of metal alloys. A fundamental question then arises: what limits the magnon lifetimes and therefore limits the magnon thermal conductivity? We are studying the exchange of thermal energy between the magnon and phonon systems as a first step toward answering that question.

We use time-domain thermoreflectance (TDTR) to measure the thermal conductivity of cuprate single crystals as a function of the frequency of thermal fields. In a time-domain thermoreflectance measurement, a laser oscillator, typically a Ti :sapphire laser operating at a 80 MHz repetition rate, is used as a pulsed source of light. The output is split into a pump and probe beam. The pump beam is modulated at a high frequency (between 1 and 20 MHz). The time of arrival of the pump and probe beams at the sample surface is adjusted by a mechanical delay line with picosecond precision. The time dependence of the temperature excursions induced by the pump provide useful information about heat capacity of thin metal films and thermal conductance of interfaces; most of the sensitivity to the thermal conductivity of the sample, however, comes from the out-of-phase response at the modulation frequency of the pump beam [1]. Approximately 10 years ago, we introduced an exact analytical solution of the diffusion equation (the analytical solution must be evaluated numerically) for an arbitrary multilayer sample in a TDTR experiment [2]. Anisotropy of high symmetry (a thermal conductivity tensor with only in-plane and through plane values) is easily incorporated. These solutions have been recently extended to the situation where the pump and probe beams are displaced with respect to each other [3].

Phenomenological two-temperature models have been used for many years to describe the coupled transport of heat by electrons and phonons in metals. Here, we apply this concept to the coupled transport of heat by magnons and phonons in the spin ladder compound Ca9La5Cu24O41. In this type of two-temperature modeling, the magnons and phonons separately satisfy a diffusion equation while the two diffusion equations are coupled to each other through a coupling parameter that has units of a thermal conductance per unit volume. Microscopically, the problem is, of course, much more complicated than this single parameter can capture. The single-parameter model will work best if the magnon occupation numbers can be approximated by a single magnon temperature and the phonon occupation by a single phonon temperature. We recently described how our conventional solution for the heat diffusion equation in the TDTR geometry can be extended to multiple channels [4].

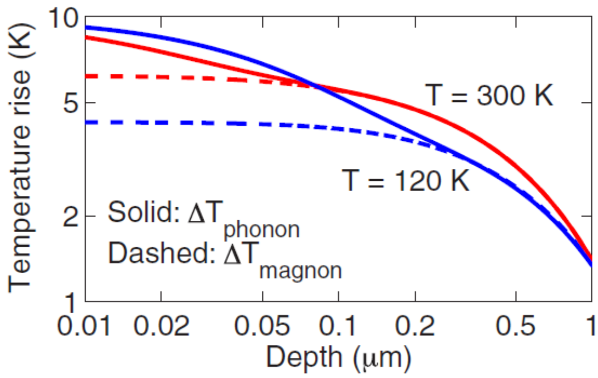

Figure 1. Model calculation of the amplitudes of the magnon and phonon temperature oscillations near the surface of a Al/spin-ladder sample during a time-domain thermoreflectance measurement with a pump modulation frequency of 10 MHz. Near room temperature, 300 K, the region of non-equilibrium is much thinner than the thermal penetration depth. At low temperatures, 120 K, the non-equilibrium region starts to overlap with the thermal penetration depth.

Because phonons can carry heat across the Al/sample interface but magnons cannot (there are no magnon excitations in the Al film transducer), the Al/sample interface creates a strong non-equilibrium between the magnon and phonon temperatures [5]. This region of non-equilibrium extends over a distance of nanoscale dimensions, approximately 50 nm at room temperature and 200 nm at 120 K, see Fig. 1. At low temperatures and high modulation frequencies, the region of non-equilibrium approaches the thermal penetration depth in the experiment and, as a consequence, the apparent thermal conductivity is strongly suppressed. We use comparisons between measured apparent thermal conductivity at different modulation frequencies and the predictions of the 2-channel model [4] to determine the magnon-phonon coupling parameter g from 80 to 300 K. Near the peak in the magnon thermal conductivity, $g \approx 10^{15}$ W m^{-3} K^{-1}, approximately two orders of magnitude smaller than a typical electron-phonon coupling parameter in a metal [5].

B. Generation of spin currents by heat currents in metallic multilayers

Cross terms of the electrical and thermal transport coefficients, i.e., the Seebeck and Peltier coefficients, have been a topic of sustained study for many decades because of their applications in sensing, solid-state cooling, and energy harvesting. There are also cross-terms that involve spin and charge currents, and cross terms for spin and heat currents. The cross-terms of spin and charge are a key topic of study in the field of spintronics. The cross-terms of spin and heat are a core consideration of an emerging discipline, often referred to as spin caloritronics.

One of the most challenging problems in the field of spin caloritronics is the detection of the spin density or spin current in a sample: the experimentalist does not have the meter for spin that is analogous to of a thermometer or a voltmeter. Often, measurements are based on the so-called inverse spin Hall effect (ISHE) where a spin current entering a normal (nonferromagnetic) metal with strong spin-orbit coupling generates an electric field that can be measured as a transverse voltage. The voltages generated by the ISHE effect are extremely small and, controversially, a susceptibile to systematic errors generated by conventional magneto-thermoelectric effects driven by heat currents flowing through the electrical contact leads. We have taken an alternative approach for detecting spin that also provides high time resolution: we detect the spin density in a normal metal using the magneto-optic Kerr effect (MOKE). By performing MOKE measurements with a pump probe apparatus, we can generate enormous heat currents (surpassing 100 GW m^{-2} K^{-1}) on picosecond time-scales and simultaneously detect spin accumulation with picosecond time resolution [6].

We are working to better constrain the calibration of the Kerr rotation as a function of spin accumulation in Cu and Au. This is a challenging process because we do not have a calibrated source of spin. Our initial experiments and analysis suggest that the Kerr rotation is determined by the strength of the spin-orbit coupling in the conduction band and is approximately 5 times stronger in Au than in Cu.

We study two types of samples that are shown schematically as Fig. 2. In the first type of sample, the spin accumulation in the normal metal is detected by time-resolved MOKE (TR-MOKE). In the second type of sample, the transfer of spin angular momentum (the so-called spin transfer torque) is detected by the amplitude of the magnetic precession that is induced into a very thin (2 nm thick) in-plane magnet made of CoFeB [6].

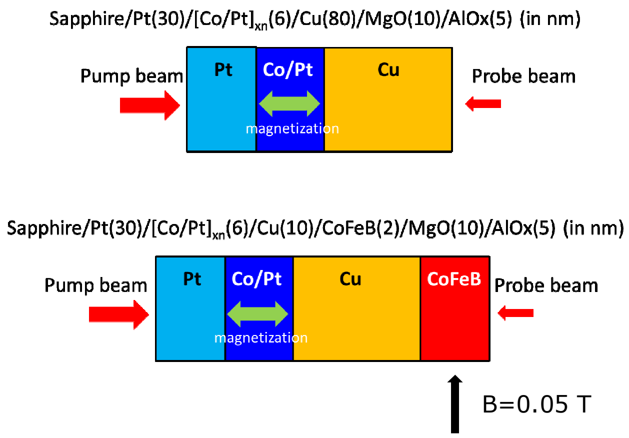

Figure 2. Schematic diagrams of the two types of samples used in the studies of thermally-driven spin currents. The numbers in the sample descriptions are thicknesses in nm. The pump beam is incident on the left and heat flows from left-to-right. (Top) Sample design for spin accumulation. The spin density in the thick Cu layer is detected by the magneto-optic Kerr effect (MOKE). (Bottom) Sample design for spin-transfer torque. The spin transfer torque is measured through the amplitude of the magnetization precession of the CoFeB ferromagnetic layer, also detected optically via MOKE.

We are working to constrain the many parameters in the models of heat transport, spin generation and spin diffusion that we use to analyze the experiments. In the initial experiments, we have found that the spin currents are predominately generated by the fast thermal demagnetization of the (Co,Pt) ferromagnetic layer. Essentially, raising the temperature decreases the equilibrium magnetization and the non-equilibrium between electrons and magnons transfer a fraction of the spin angular momentum to the conduction electrons which then diffuse into the adjacent layers. A smaller amount of spin current is generated by the spin-dependent Seebeck effect (SDSE) of the ferromagnetic layer. The SDSE is due to the fact that the product of the Seebeck coefficients and conductivities of the up and down spin sub-bands are not equal; therefore, a heat current passing through a ferromagnet produces a spin accumulation near the surfaces or interfaces of the ferromagnetic layer that can diffuse into adjacent layers [6].

We argue that the experimental design illustrated by Fig. 2 will provide a rich platform for studies of the coupling of heat and spin in metallic multilayers. With some advances, we will soon have calibrated sources of spin and calibrated detectors of spin, both with picosecond time resolution. This platform can be used to quantitatively study thermal generation of spin currents by the spin-dependent Seebeck effect and ultrafast demagnetization, as well as transport physics of spin such as the transport and mixing of spin at interfaces.

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