Magnon scattering in the transport coefficients of CoFe thin films

S. Srichandan, M. Kronseder, S. Wimmer*, M. Vogel, C. Back, H. Ebert*, <u>C. Strunk</u>

Experimental and Applied Physics, Regensburg University *Dept. of Chemistry, Ludwig-Maximilian University, Munich



Spin - caloritronics

many new types of spin-dependent transport effects:

• spin-Hall and inverse spin-Hall effects

Kato et al., Science 2004; Wunderlich et al., PRL 2005; Saitoh et al., APL 2006

- transverse and longitudinal spin-Seebeck effects Uchida *et al.*, Nature 2008; Jaworski *et al.*, Nat. Mat. 2010; Uchida et al., Nat. Mat. 2010
- spin-Peltier and spin-Nernst effects

Flipse, et al., Nat. Nanotech. 2012; PRL 2014; Meyer et al., Nat. Mat. 2017

sometimes an effect can disappear again...

M. Schmid, S. Srichandan, C. S., C. Back et al., PRL 2013

how well do we understand the more conventional transport phenomena ?

- spin-dependent resistivity ?
- spin-dependent thermopower ?
- spin-effects in thermal conductance ?

Magnon scattering in itinerant ferromagnets

different types of quasi-particles contribute to and affect transport of charge, spin and heat in ferromagnets:

- electrons with majority and minority spins
- phonons
- magnons

studied in the 1950's – 1970's, but no detailed understanding at that time

- time to take a fresh view:
- precise role of spin-degrees of freedom?
- systematic change of electron density as control parameter
- comparison to state-of-the-art model calculations

Can we separate the effects from different sub-systems?

Device fabrication



Al contacts to measure resistivity and thermopower

SiN based suspended microcalorimeter



thermometer

XRD characterization of CoFe films:

- x_{co}= 0. 22, 0.22: single **bcc** phase,
- x_{co}=0.36, 0.7: mixed **bcc + fcc** phase

see also A. D. Avery, et al., PRL 2012

Electrical resistivity



- monotonic decrease with decreasing electron density
- temperature dependent part: Δρ=ρ(296 K)-ρ(25 K) ~ 2-5 μΩ cm, comparable to other experimental group¹.
- Δρ smaller than the Δρ of bulk
 Fe² (10.41 μΩ cm) and of Co³
 (6.23 μΩ cm), respectively.

¹C. Ahn *et al., Jour. Appl. Phys.* **108**, 023908 (2010)
²M. Rubinstein *et al., Phys. Rev. B* **37**, 15 (1988)
³J. W. C. De Vries *et al., Thin Solid Films* **167**, 25-32 (1988)

Resistivity as a function of Co concentration



¹M. Rubinstein *et al., Phys. Rev. B* **37**, 15 (1988) ²J. W. C. De Vries *et al., Thin Solid Films* **167**, 25-32 (1988) ³P. P. Freitas *et al., Phys. Rev. B* **37**, 6079 (1988)

- besides chemical disorder strong contribution from grain boundary scattering
- violation of Matthiessen's rule



Understanding the effects of different scattering processes

- measure *T*-dependence of different transport coefficients while systematically tuning the Fermi level through the (pseudo)band structure
- comparison to microscopic theory (Ebert group, LMU Munich)
 - For band structure calculation, Munich SPR-KKR-program package has been used. This includes the fully relativistic Korringa-Kohn-Rostoker multiple scattering Green's function formalism to evaluate Kubo'sformula.

H. Ebert *et al., Rep. Prog. Phys.* **74**, 096501 (2011)

2. Temperature dependent calculation for resistivity using **alloy-analogy model** (AAM) based on coherent potential approximation (CPA) **including vertex corrections**.

H. Ebert et al., Phys. Rev. B 91, 165132 (2015)

Thermopower from generalized Mott model using temperature dependence from electrical conductivity and Fermi-Dirac distribution.

S. Wimmer et al., Phys. Rev. B 88, 201108 (R) (2013); Phys. Rev. B 89, 161101 (R) (2014)

Band structure (Bloch spectral function)



- red lines: shape of d-like band of the spin-up channel
- 0 represents the Fermi energy E_F
- E_F inreases with respect to bands when electron density increases

S. Wimmer *et.al.* (2016)

Separation into electron-phonon and electron-magnon scattering contributions



- phonon and magnon energy scales differ by a factor 10
- electron-magnon scattering contribution is small

M. V. Kamalakar *et al.,* Appl. Phys. Lett. **95**, 013112 (2009)

 chemical and vibronic disorder scattering contribution in calculations underestimates the residual resistivity and overestimates the temperature dependence

S. Wimmer *et.al.* (2016)

Magnetic contribution to resistivity remaining after subtraction of Bloch-Wilson fit (?)



Thermopower S(T)



- S(T) is mostly negative in sign.
- monotonic increase with decreasing electron density.
- visible change of sign of curvature at lower temperatures.

Separation of Mott-like and magnon drag contributions



from Watzman et al.

low energy magnon number ~ $T^{3/2}$

70 % Co

Separate magnon drag and Mott-like contributions



- maximum S_{magnon-drag} is 15 μV/K at 296 K for film with x_{Co}=0.2.
- S_{magnon-drag} changes sign from negative to positive following sign of bulk Co and Fe

S. J. Watzman et.al., Phys. Rev. B 94, 1444 (2016)



- conjectured S_{Mott} extracted from experiment is linear in T by construction.
- thermoelectric parameter $\eta = -3eS_{Mott}T_F/\pi^2k_B = 5.2$ for film with $x_{Co}=0.2$ is comparable with $\eta=7.6$ for liquid Fe

K. Hirata et al. J. Phys. F: Metal Phys. 7, 3 (1977)

origin of the sign change of magnon drag thermopower?



S_{magnon-drag} may contain information on the spin transfer torque parameters



Y. Tserkovnyak et al., J. Magn. Mag. Mat. 320, 1282 (2008)

Mott-like contribution compared to microscopic calculation



 Mott-like contribution to thermopower extracted from phenomenological decomposition



 Calculation using generalized Mott relation shows band structure effects.

$$S = -\frac{1}{eT} \frac{\int \sigma(\varepsilon)(\varepsilon - \mu) \frac{-\partial f(\varepsilon, \mu, T)}{\partial \varepsilon} d\varepsilon}{\int \sigma(\varepsilon) \frac{-\partial f(\varepsilon, \mu, T)}{\partial \varepsilon} d\varepsilon} d\varepsilon$$

Residual thermopower



- $S_{residue}(T) = S S_D S_M$ is (much) smaller than S_M and S_D
- For films with $x_{co} = 0.2$ and 0.22,

S_{residue} is similar to phonon drag in bulk Ni (and Al reference leads)

 For films with x_{co}= 0.36, 0.5 and 0.7, effective in-elastic scattering and phonon drag effect

Thermal platform model for thermal conductance



SiN based suspended microcalorimeter



Comparison with microscopic calculation results



- theory reproduces nicely the concentration dependence
- numbers are a factor two larger values of κ
 most probably connected to underestimation of ρ in theory
- theory works better than for ρ and S.

Lorenz ratio and violation of Wiedemann-Franz law



- Lorenz number L(T) / $L_0 = \kappa(T) \rho(T) / T$
- L > L₀: For films with x_{co}=0.2 and 0.22 due to the significant phonon (magnon?) contribution to thermal conductivity (no fcc phase precipitation)
- L < L₀: For films with x_{Co} =0.36, 0.5 and 0.7 due to enhanced inelastic scattering of electrons with phonons and magnons (with fcc phase precipitation)

Conclusions

- Experimental platform enables study of all transport coefficients of the very same film
- Theory significantly underestimates composition dependent resistivity
- Thermopower is phenomenologically decomposed into Mott-like and Magnon drag terms the latter changes sign for large Co-concentrations
- Thermal conductivity is mainly electronic
- Significant deviations from Wiedemann-Franz law observed in experiment.

