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Spin-pumping-enhanced magnetic damping in ultrathin Cu(0 0 1)/Co/Cu and Cu(0 0 1)/Ni/Cu films

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ABSTRACT

The influence of the Cu capping layer thickness on the spin pumping effect in ultrathin epitaxial Co and Ni films on Cu(0 0 1) was investigated by *in situ* ultrahigh vacuum ferromagnetic resonance. A pronounced increase in the linewidth is observed at the onset of spin pumping for capping layer thicknesses d_{Cu} larger than 5 ML, saturating at d_{Cu} = 20 ML for both systems. The spin mixing conductance for Co/Cu and Ni/Cu interfaces was evaluated.

1. Introduction

The structure and magnetic properties of ultrathin Cu(0 0 1)/Co/Cu and Cu(0 0 1)/Ni/Cu films have been studied extensively in past years [1–5]. Their tailorable magnetic performance makes such systems excellent candidates for fundamental investigations of magnetization dynamics. One of the most important features for the magnetic performance of devices for spintronics applications such as fast high density magnetic storage devices, spin valves, etc. [6], is the rate at which the magnetization can be switched, e.g. in order to write a bit of information. The magnetization dynamics in the ultrathin film regime, where the influence of bulk properties is eliminated, allows the direct observation of interfacial effects.

A precession of the magnetization in the ferromagnetic (FM) layer, i.e. Co or Ni, causes a spin current, which propagates through the interface into the nonmagnetic (NM) Cu substrate as well as into the cap layer, which act as spin sinks [7–10]. This spin pumping enhances the intrinsic relaxation of the magnetization in the FM layer, thus making switching processes faster.

FMR is a powerful method used for the determination of magnetic anisotropies and the characterization of magnetization dynamics. The utilization of FMR *in situ* in ultrahigh vacuum (UHV) allows for direct step-by-step preparation and measurement of the systems, which are thus characterized by high structural ordering and material purity, otherwise not achievable. In this work UHV FMR studies have been performed on single crystalline ultrathin Co and Ni films on Cu(0 0 1) with Cu cap layers of variable thickness, which are much thinner than the spin diffusion lengths of these systems [11]. In this thickness regime the interface-related relaxation mechanisms like spin-pump effect are dominant [12], especially at first contact between the vacuum side of the FM film and the NM cap layer.

The precessional motion of the magnetization vector $\vec{M}$, which for ferromagnetic films is regarded as a macro-spin, can be described by the Landau–Lifshitz–Gilbert (LLG) equation of motion [13,14]:

$$\frac{\partial \vec{M}}{\partial t} = -\gamma(\vec{M} \times \vec{H}_{\text{eff}}) + \frac{\alpha}{M_S}\left(\vec{M} \times \frac{\partial \vec{M}}{\partial t}\right), \tag{1}$$

where $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio, M_S the saturation magnetization. $\vec{H}_{\text{eff}}$ is the effective field including the external static field and the internal fields, i.e. anisotropy fields and microwave field, respectively. α is the dimensionless parameter of the intrinsic Gilbert damping, which depends on the strength of the spin–orbit coupling $\lambda_{LS} : \alpha \rightarrow \alpha_G \propto \lambda_{LS}^2$ [15]. Gilbert damping can be understood in terms of a viscous force, where the coupling of the spins to the orbital motion acts as a restoring force leading to a spiraling down of the spins toward the direction of the external field $\vec{H}_0$. This relaxation process is usually dominant in most cases and is called 'intrinsic'. However, in further examining the system the term 'intrinsic' looses its meaning at thicknesses where almost no volume contribution exists.

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Other significant relaxation processes could be (i) eddy current damping, (ii) phonon dragging, and (iii) spin-pumping. (i) and (ii) can be neglected for ultrathin film systems since the damping amplitude depends on the thickness of the film $\alpha_{\text{eddy}} \propto d^2$ [16] and the phonon dragging contribution is an order of magnitude smaller than the Gilbert damping α_G [17,18]. Thus the only other significant additional contribution in this thickness range is spin pumping, which can be regarded as a Gilbert-like damping, since it has a similar mathematical form in the LLG [19]. Along the *in plane* easy axis the relation of the peak-to-peak linewidth ΔH_{pp} of the FMR signal to the microwave frequency is [20,21]

$$\Delta H_{pp} = \alpha_{\text{eff}} \frac{2}{\sqrt{3}} \frac{\omega}{\gamma} + \Delta H^0_{pp}, \tag{2}$$

where α_{eff} is the sum of contributions to the relaxation and ΔH^0_{pp} is the contribution of inhomogeneities, which is constant for all frequencies and geometries. Furthermore, all films studied in this work were measured at 9 GHz. Prior *in situ* FMR experiments by Platow et al. on ultrathin Ni on a limited frequency range [22] give no indication for a two-magnon scattering contribution [23] to the damping. This leaves a total damping process involving only the Gilbert damping and spin-pumping:

$$\alpha_{\text{eff}} = \alpha_G + \alpha_{\text{pump}}. \tag{3}$$

However, note that two-magnon scattering would lead to an overestimation of α_{eff}. The study of the spin-pump effect on capped single films requires systems with well defined interfaces [12,24]. Cu/Co/Cu and Cu/Ni/Cu systems represent a good case for even interfaces due to the very small mismatch of the lattice constants, which is 1.9% for Co and 2.5% for Ni [25], and their pseudomorphic growth.

Precession of the magnetization vector around an effective field axis, like in FMR, is known to generate a spin current $\vec{I}_{\text{pump}}$, which flows from the FM layer into the NM layer [7]. If the NM layer is thick enough the spin current is dissipated there by spin-flip processes. Thus, torque is carried away from the precession and reduces the precessional energy of the FM layer. This process can be regarded as another damping mechanism. The contribution of spin pumping to the overall relaxation can be derived from the conservation of angular momentum in the FM layer [26,27]:

$$\frac{1}{\gamma} \frac{\partial \vec{\mu}_{\text{tot}}}{\partial t} = \vec{I}_{\text{pump}} \Rightarrow \frac{d\vec{m}}{dt} = \frac{\gamma}{M_S V} \vec{I}_{\text{pump}}, \tag{4}$$

where $\vec{\mu}_{\text{tot}} = \vec{m} M_S V$ is the total magnetic moment and $\vec{m}$ is the unit vector of magnetization. The spin current $\vec{I}_{\text{pump}}$ flowing through the FM/NM interface can be written as [7,19]

$$\vec{I}_{\text{pump}} = \frac{\hbar}{4\pi} \left[g_r^{\uparrow\downarrow} \left(\vec{m} \times \frac{d\vec{m}}{dt} \right) - g_i^{\uparrow\downarrow} \frac{d\vec{m}}{dt} \right], \tag{5}$$

where $g_r^{\uparrow\downarrow}$ is the real part of the spin mixing conductance $g^{\uparrow\downarrow}$. The imaginary part $g_i^{\uparrow\downarrow}$ can be neglected since $g_r^{\uparrow\downarrow} \gg g_i^{\uparrow\downarrow}$ [28–30]. Hence, inserting Eq. (5) into (4) results in

$$\frac{d\vec{m}}{dt} = \frac{\gamma}{M_S V} \frac{\hbar}{4\pi} g^{\uparrow\downarrow} \left(\vec{m} \times \frac{d\vec{m}}{dt} \right), \tag{6}$$

representing the additional loss of angular momentum from the FM layer. Adding the right hand side to the general LLG equation (1) rewritten in terms of the unit vector of magnetization yields

$$\frac{d\vec{m}}{dt} = -\gamma \left(\vec{m} \times \vec{H}_{\text{eff}} \right) + \underbrace{\left(\alpha_G + \frac{\gamma}{M_S V} \frac{\hbar}{4\pi} g^{\uparrow\downarrow} \right)}_{\alpha_{\text{eff}}} \left(\vec{m} \times \frac{d\vec{m}}{dt} \right). \tag{7}$$

Comparing Eqs. (3) and (7) one arrives at the expression for the spin-pumping contribution to the damping:

$$\alpha_{\text{pump}} = \frac{\gamma}{M_S V} \frac{\hbar}{4\pi} g^{\uparrow\downarrow} = \frac{g \mu_B}{4\pi M_S d_{FM}} \frac{g^{\uparrow\downarrow}}{S}, \tag{8}$$

where d_{FM} is the thickness of the ferromagnetic layer and S is the interface area.

This expression describes the damping caused by the total loss of spin energy transported by $\vec{I}_{\text{pump}}$, which is the case of having an ideal spin sink, i.e. no backflow due to spin accumulation in the NM cap layer. For a complete approach, one has to take this backflow $\vec{I}_{\text{back}}$ into account, which depends on $\vec{I}_{\text{pump}}$, the thickness of the NM cap layer d_{Cu}, and the spin diffusion length l_{sd} of about 350 nm in Cu at room temperature [11]. The net spin current $\vec{I}_{\text{net}} = \vec{I}_{\text{pump}} - \vec{I}_{\text{back}}$ equates to [12,29]

$$\vec{I}_{\text{net}} = \left[1 + \frac{\tau_{sf} g^{\uparrow\downarrow}}{hNSl_{sd} \tanh(d_{Cu}/l_{sd})} \right]^{-1} \vec{I}_{\text{pump}}, \tag{9}$$

where N is the density of states per spin and τ_{sf} the spin flip time. This can be expressed in terms of α_{pump} by comparing Eqs. (5), (6), and (8). Using N of a free electron gas, $N = (m_e k_F)/(2\pi^2 \hbar^2)$ and $l_{sd} = \hbar k_F / m_e \sqrt{\tau_{el} \tau_{sf}/3}$, with the electron mass m_e, we arrive at a final expression for the damping parameter of the spin-pumping contribution:

$$\alpha_{\text{pump}} = \frac{g \mu_B}{4\pi M_S d_{FM}} \frac{g^{\uparrow\downarrow}}{S} \left[1 + \frac{\pi\sqrt{3}}{k_F^2 \sqrt{\varepsilon}} \frac{g^{\uparrow\downarrow}}{S} \frac{1}{\tanh(d_{Cu}/l_{sd})} \right]^{-1}, \tag{10}$$

where $\varepsilon = \tau_{el}/\tau_{sf}$ is the spin flip probability per scattering event, with τ_{el} being the elastic scattering time, and k_F the Fermi wave vector, $k_F = \sqrt{2 m_e E_F}/\hbar$ which is known to be $k_F = 1.36 \times 10^8\ \text{cm}^{-1}$ for bulk Cu. The free parameters of this model are hence the conductance per unit area $g^{\uparrow\downarrow}/S$, the arbitrary spin flip probability ε and the spin diffusion length l_{sd}, which have been determined for this system, however, not been tested at length scales of a few monolayers and at first contact of a cap layer. Thus by measuring the FMR linewidth ΔH_{pp} of single uncapped Co and Ni films and gradually adding Cu we can calculate the change in α_{pump} using Eqs. (2) and (3) while assuming $\alpha_G = \text{const.}$, and determine the fit parameters for each system by applying the model in Eq. (10) for the thickness dependence.

2. Experimental details

The thin films examined in this work were grown *in situ* in UHV with a base pressure of 5×10^{-11} mbar on a Cu(0 0 1) single crystal disc, 5 mm in diameter and 2.5 mm thick. Prior to film deposition, the substrate was sputtered with Ar^+ ions at a partial pressure of 5×10^{-5} mbar with 3 keV for 10 min, then annealed at 850 K and sputtered again with 1 keV for another 10 min to smoothen the surface. After a final annealing again at 850 K for further 10 min, the Co (Ni) films with thicknesses of 1.6, 1.7, and 1.8 (6.0 and 7.0) monolayers (ML), respectively, were grown from high purity targets by means of electron beam evaporation. The optimum growth rate for all films was found to be 1 ML/min. The thickness was controlled during evaporation by means of medium energy electron diffraction (MEED). After deposition all films were annealed at 420 K for 10 min in order to smoothen the surface.

The Cu cover layers were prepared by the same procedure. However, no MEED signal could be detected for thicker films. Therefore, the thickness of the Cu cover layer was calculated according to the exposure time, based on a series of calibration runs on 10 ML thick Co films. The samples were annealed again at 420 K for 10 min after each additional Cu deposition step.

In situ FMR measurements were performed at 8.87 GHz using a cylindrical cavity with the TE_{012} mode. The cavity was mounted around the outside of a UHV quartz glass finger to the UHV chamber. This enables the measurement of FMR spectra *in situ* in UHV directly after preparation, hence without breaking the vacuum. Also it allows to change the thickness of the cap layer