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Synthetic route to low damping in ferromagnetic thin-films

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ABSTRACT

Previous theory indicated that the individual monolayers within transition metal ferromagnet thin-films contribute different magnitudes to the total ferromagnetic damping. Here, the aim was to investigate if the thin-film damping could be reduced by electronic engineering of the higher damping regions via localized doping. We present new theoretical analysis and experimental results for sputtered Co thin-films in which the upper and lower surface regions were locally doped with Cr. Theory indicates that local doping does reduce the damping and the experiments show a comparable reduction of the damping with increasing local doping up to 30% Cr, while the measured damping falls further with higher local doping, which may be attributed to changes in the film structure. This work opens a route to create low-damping magnetic thin-films.

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Damping of precessional magnetization is fundamental to the processes of magnetization change in magnetic materials and critical for device performance in magnetic technology, with ultra-low damping in thin-films a significant driver for spintronic and magnonic applications. While the physical basis of damping is established,^{1–4} it is only recently that theory has shown quantitative agreement with measurements in ferromagnet alloys⁵ and that the density of states at the Fermi level has been mapped out for the compositions of metallic, half-metallic, and oxide ferromagnetic thin-films with ultra-low damping.^{5–9} This composition dependence has been one critical route to ultra-low damping. Elsewhere, theory indicates that in ferromagnetic films the largest contributions to damping often arise from the outermost atomic layers.¹⁰ Here, it was realized that modifying the electronic structure of only these outer layers experimentally could significantly lower the damping in a wide range of alloys and we demonstrate this “synthetic” route to creating ultra-low damping ferromagnetic layers by synthesizing Co thin-films where the upper and lower few atomic layers are locally modified by doping. The damping is reduced by more than an

order of magnitude. We explain these results theoretically through changes to the electronic states of the locally doped surface layers. The important role of damping in magnetization processes of ferromagnetic materials was recognized by Landau and Lifshitz¹¹ in the 1930s, and both theoretical and experimental research attention to damping has continued since that time. The functional benefits of low damping are widely recognized in spintronics, for example, enabling low current-density spin torque transfer switching,¹² while in magnonics, the larger magnon lifetimes that enable longer mean free path lengths also require ultra-low damping.¹³ Low damping research has often focused on the magnetic insulators such as yttrium-iron-garnet (YIG),^{14,15} which is suitable for magnonics, while studies of ultra-low damping conductive ferromagnetic thin-films for spintronics has included works on the half-metallic perovskite $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_{0.7}$ (LSMO)⁹ and Heusler alloys Co_2FeAl ⁶ and NiMnSb .¹⁶

Despite the ultra-low damping reported for these materials, their applications in spintronics, spin-orbitronics and magnonics, and their integration with complementary metal-oxide semicon-

ductors (CMOS) devices present major challenges. The lack of electronic conductivity and critical issues with the material preparation conditions present major problems for the application of these materials. Hence, the continued interest in understanding and tuning the damping in thin-film ferromagnetic transition metal alloys, as these materials have benefits in terms of their electronic conductivity and ease of fabrication with conventional methods. Significantly, first principles calculations by Mankovsky *et al.*⁴ detailed the variations of the Gilbert damping in transition metal ferromagnets, including the compositional dependence of damping in the $\text{Fe}_{1-x}\text{Co}_x$ alloy series. This inspired a detailed study by Schoen *et al.*⁵ that explored experimentally this compositional dependence, demonstrating ultra-low damping with 25% cobalt in $\text{Co}_x\text{Fe}_{1-x}$ and explained theoretically the origin of the ultra-low damping in terms of the density of states at the Fermi energy and intraband scattering, which has a minimum in the CoFe series for 25% cobalt. Overall, this understanding of damping in transition metals highlights the limited range of conventional ferromagnetic alloys with low damping.

Alternative routes to low damping would expand the range of ferromagnetic materials for applications. Experiments have shown a small reduction in damping through bulk doping of $\text{Ni}_{81}\text{Fe}_{19}$ with a small percent of Cr and Ag,¹⁷ while a small reduction in damping was obtained by ion-beam-induced interfacial intermixing in $\text{Ni}_{81}\text{Fe}_{19}/\text{Cr}$ bilayer thin-films,¹⁸ although work on ion implanted Cr in $\text{Ni}_{81}\text{Fe}_{19}$ showed a dramatic increase in damping for the implantation of 4% Cr.¹⁹

A detailed theoretical study by Barati *et al.*¹⁰ on damping in magnetic thin-films calculated the individual monolayer contributions to the total damping in Fe, Co, and Ni thin-films.¹⁰ This showed that for Fe and Co the outer monolayers make a very large contribution to the damping, while for Ni the variation through the thickness was much smaller.

Here, we have experimentally and theoretically investigated locally modifying the electronic structure of the outer atomic layers of transition metal ferromagnetic thin-films by doping as a synthetic route to creating conductive thin-films with reduced ferromagnetic damping. Co was selected as the ferromagnet since theory shows that the highest damping is associated with several of the outermost monolayers¹⁰ at each surface, although in principle the methodology is applicable to Fe and transition metal alloys. Modifying the electronic structure of the outer most layers was achieved by locally doping the Co film at the beginning and the end of the film deposition process. Cr was selected as the local dopant based upon the experimental results of bulk doping by Rantschler *et al.*¹⁷ and interfacial intermixing reported by King *et al.*¹⁸ Furthermore, Cr was selected as the addition of Cr to both hcp and fcc Co maintains the original crystallographic structure up to at least 30% Cr with a limited increase in the lattice parameter.²⁰

Co films of thickness ~ 5 nm (50 \AA) were prepared by magnetron sputtering at room temperature onto oxidized silicon substrates and capped with ~ 3 nm Cu to protect the Co. Local doping of the upper and lower surface regions of the Co films was introduced by calibrated co-sputtering of Cr at the beginning and the end of continuous Co deposition. The quantity of Cr doping was controlled by the sputter deposition rate. X-ray reflectivity (XRR) measurements and analysis using the differential evolutionary

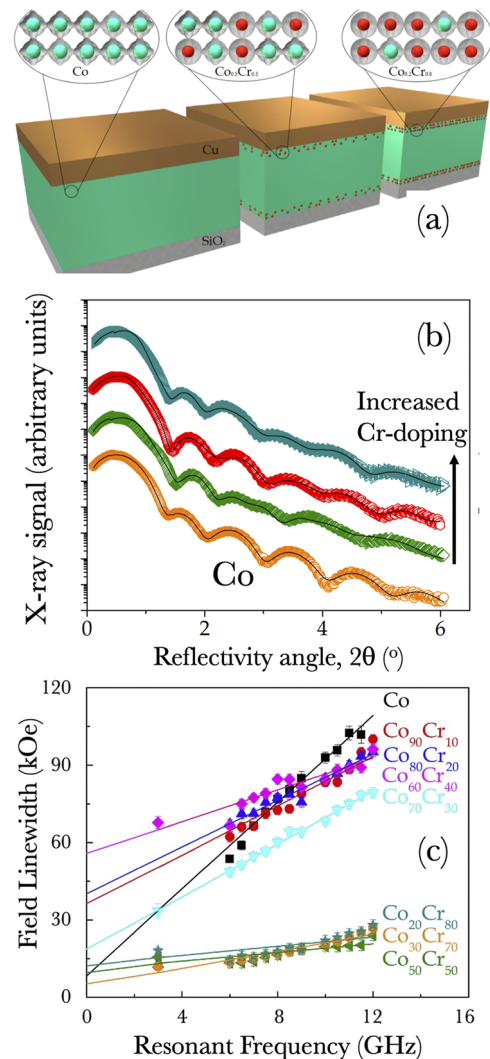


FIG. 1. (a) Illustration of the Cu capped ferromagnetic Co thin films locally doped in the upper and lower regions with Cr that form the basis of this study. (b) Examples of the x-ray reflectivity and best-fitting GenX code simulations for a pure Co film and films where the upper and lower regions are doped with increasing levels of Cr. The XRR curves are offset on the y-axis for presentation. (c) Resonance linewidths as a function of the resonant frequency for Co films with different levels of Cr doping in the upper and lower regions.

algorithm code GenX²¹ indicates a Co thickness of 47–48 Å with 2–3 Å thick regions of CoCr at the upper and lower surfaces and a 35 Å Cu capping layer. The approximate film structure of the locally doped thin-films was therefore $\text{SiO}_2/\text{Co}_{1-x}\text{Cr}_x$ (2.5 Å)/Co (47.5 Å)/Co_{1-x}Cr_x (2.5 Å)/Cu (35 Å), where the Cr doping of the upper and lower Co surface regions, x , ranged from 0% to 80% Cr. Figure 1(a) illustrates the thin-film structure with different Cr doping concentrations in the upper and lower regions of the Co film and Fig. 1(b) shows the measured XRR for selected samples and the best-fitting simulated results.

For comparative purposes, a series of uniformly doped Co films was also prepared with the structure $\text{SiO}_2/\text{Co}_{1-x}\text{Cr}_x$ (50 Å)/Cu (30 Å), where x ranged from 0% to 8% Cr.

Magnetic damping was determined from ferromagnetic resonance (FMR) spectroscopy. Measurements were made using a RF generator and microwave diode/lock-in amplifier detection method in the magnetic field domain. The magnetization and the saturating magnetic field were in the film plane. This method allows extraction of the Gilbert damping parameter from the field (H) dependent resonant line-widths ΔH using

$$\Delta H = \Delta H_0 + \frac{4\pi f}{\gamma} \alpha. \quad (1)$$

Analysis of the field dependent measurements provides a direct method to obtain the Gilbert damping parameter, α , and the inhomogeneous line-width, ΔH_0 , and the benefits of this approach have been previously reported.²² However, it is recognized that two-magnon scattering may be contributing to this apparent Gilbert damping, but cannot be resolved without going to higher resonant frequencies, where curvature would be observed,²³ so it is appropriate here to refer to the Gilbert damping parameter as the effective damping parameter.

Figure 1(c) shows examples of the experimentally measured resonance line-widths as a function of the resonant frequency for Co films with different levels of Cr doping within the upper and lower few monolayers, the figure also shows the linear best fits, from which the effective damping parameter and inhomogeneous line-broadening contributions were determined.

The 3 nm Cu cap is likely to be partially oxidized, reducing the thickness of the Cu metal. This may increase scattering and damping via spin-pumping into the Cu, although this is a small effect in Cu.²⁴ The raw damping thus also include a small contribution from spin-pumping into the Cu, α_{sp} ²⁵ and from the inductive interaction between the precessing magnetization and the waveguide, termed radiative damping,^{26,27} α_{rd} , both of which have been considered here.

The effective damping for the un-doped Cu capped Co film here was determined to be 0.0115, which is close to the values reported by Oogane *et al.*²⁸ and Azzawi *et al.*²⁹ and larger than values obtained by others.^{5,30} These variations may be associated with differences in the film microstructure resulting from the deposition and the underlayer/substrate used, as this influences the local electronic environment that ultimately determines the damping, which for Co includes hcp and fcc phases. For the uniform Cr-doped Co films, a reduction in damping down to 0.0080 was observed with uniform Cr doping up to 8%.

Figure 2(a) shows the change of the effective damping as a function of the Cr doping concentration in the upper and lower few atomic layers of the Co films. Figure 2(a) also shows the resulting effective damping after subtracting simplified estimates to indicate the magnitude and sense of the combined effects of spin-pumping and radiative damping contributions, based on the work of Tokaç *et al.*³⁰ and Schoen *et al.*⁵ The work of Tokaç *et al.* indicates a spin pumping contribution for Co(5 nm)/Cu(3 nm) of order 0.003, while the radiative contribution to damping determined by Schoen *et al.* for higher moment CoFe alloys was around 0.0005, and using these values, a conservative correction of 0.0015 was used.

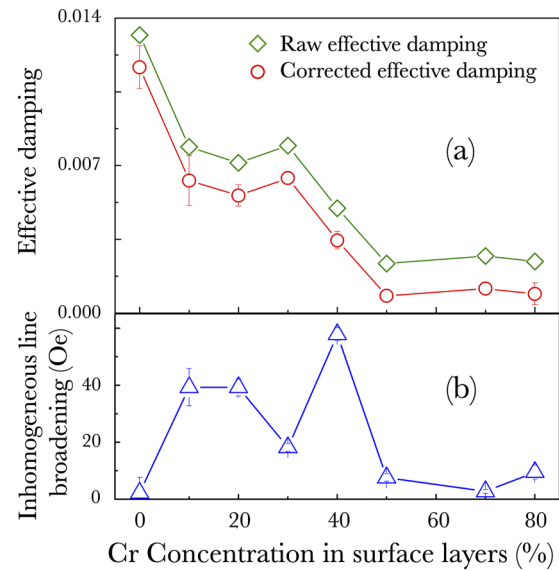


FIG. 2. (a) The measured effective damping and the effective damping after correction for spin-pumping and radiative contributions, as a function of the local Cr doping concentration in the upper and lower few monolayers of Co thin-films. (b) The inhomogeneous line broadening, as a function of the local Cr doping concentration in the upper and lower monolayers of Co thin-films.

The spin-pumping is small, but could vary with the doping at the interface, which is not accounted for in the simple correction. While the corrections are small and do not affect the trend in the damping, the damping values are very low and correcting for these effects provides a better indication of the low effective damping of a magnetic layer.²⁷

With Cr doping of the upper and lower regions of the Co films, the damping falls significantly with 10% Cr, then remains constant up to 30% and falls further between 30% and 50% Cr doping, reaching a lowest damping value below 1×10^{-3} . It is interesting to note that this effect does not follow a capping layer thickness effect, where only an increase of the damping was observed.²⁹

Figure 2(b) shows the inhomogeneous line broadening, ΔH_0 , as a function of Cr doping in the upper and lower Co atomic layers. This additional line broadening is physically associated with overlapping FMR responses arising from magnetic inhomogeneities that increase the overall line-width through additional scattering processes.³¹ With Cr doping of the upper and lower surfaces, this additional linewidth broadening is very low for the undoped Co and when the upper and lower surfaces are doped above 40% Cr. Between 10% and 40%, the extrinsic damping is increased, but without a clear trend.

To understand the physical basis for the reduction of Gilbert damping of Co thin-films with Cr doping within a few atomic layers at the top and the bottom of the films, theoretical calculations were made on an idealized atomic layered structure. The experimental setup was modeled by an unsupported epitaxial slab of 20 monolayers of fcc Co capped with four monolayers of fcc Cu. XRR analysis of the samples indicates that Cr impurities are

primarily located in the outer few monolayers layers of Co, but this cannot be better defined. The calculation of the Gilbert damping was performed using the layer dependent version of the Kamberský formula derived in Ref. 10 that was extended to include Cr doping. Details of our method of calculation and the bulk parameters used are found in Ref. 10, where the overall damping is the sum of the individual layer contributions. This approach is known to give agreement between theory and experiment for a variety of layered systems,²⁹ despite differences between the theoretical and experimental atomic structures. For a pure Co film, the theory shows that the monolayer contributions to the damping are dominated by the outer layers as reported previously.¹⁰ To simulate the Cr doping in this work, we modified the electronic potentials of Co. In particular, for a doping density, ρ_{Cr} , of Cr atoms on any given layer of Co, we performed the following: an appropriate shift in the on-site potentials so as to maintain charge neutrality; a linear scaling of the bulk Co exchange splitting $\Delta = \Delta_{\text{Co}}(1 - \rho_{\text{Cr}})$, to reflect the reduction in the magnetic moment produced by Cr; a linear change in the bulk Co spin-orbit coupling parameter $\xi = \xi_{\text{Co}}(1 - \rho_{\text{Cr}}) + \xi_{\text{Cr}}\rho_{\text{Cr}}$; an increase in the electronic broadening Γ , to reflect the scattering effect of Cr doping. The latter was calculated by an elementary model assuming that the broadening is proportional to the scattering probability, i.e., to the Cr doping density, such that $\Gamma = \Gamma_{\text{Bulk}} + k_s\rho_{\text{Cr}}$. Since the exact contribution of Cr scattering to broadening is not known, we treated both Γ_{Bulk} and k_s as empirical parameters. Values of $\Gamma_{\text{Bulk}} = 0.00212$ eV and $k_s = 0.05$ eV were found to give a good fit to the experimental results for $\rho_{\text{Cr}} = 0\%$ and 10%.

Figure 3 shows the calculated effect of doping the outer layers on the intrinsic damping of Co doped with Cr within the atomic layers at the top and the bottom of the films. This indicates that the addition of a few 10s of % Cr doping in the outer monolayers significantly reduces the damping contributions of these outer layers and hence reduces the overall damping of the Co thin-film system. Beyond 30% Cr doping, there is little further change in the damping.

Figure 4 provides a comparison of the experimentally determined effective damping with the theoretically derived total Gilbert damping as a function of Cr doping. The bulk value of the scattering parameter was taken to be $\Gamma_{\text{Bulk}} = 0.0021$ eV, which gives good agreement with experiment for a pure Co film (0% Cr doping). The value of the scattering constant was chosen to be $k_s = 0.05$, which approximately aligns the theory and experiment data for 10% Cr doping. We considered two theoretical scenarios: one with Cr doping confined to the outer two layers (dashed line) and another that allowed $0.1\rho_{\text{Cr}}$ to diffuse into the interior (solid line). For the latter case, we solved the diffusion equation to obtain the Cr distribution, the outcome of which is that almost all the Cr was confined to the outer three or four monolayers.

In both cases, theory and experiment are in agreement for Cr concentrations up to 30%, where the reduction in the Gilbert damping is theoretically attributed to a reduction of the intraband electronic transitions in the doped boundaries layers, which, since these monolayers initially contribute most to the damping, reduces the total film damping significantly. At higher concentrations there is a further sharp decline in the experimental values of the effective damping that is not observed in either of the two theoretical models. These observations may be explained in terms of

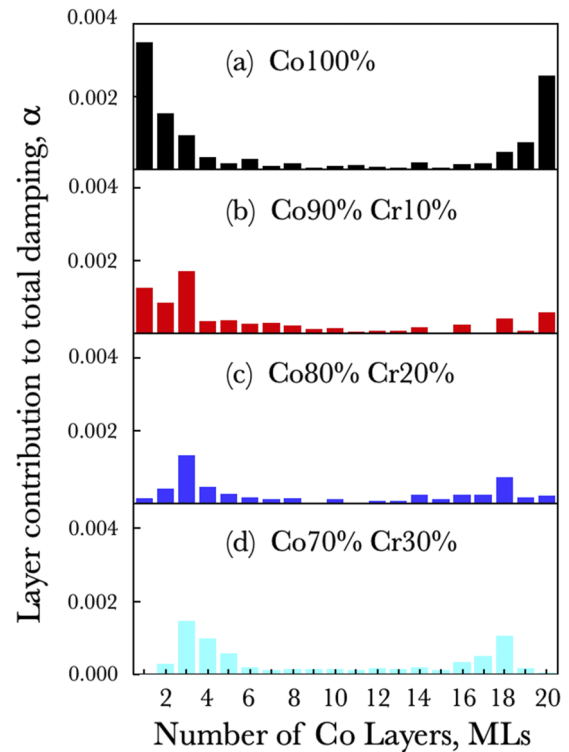


FIG. 3. Theoretical analysis showing the individual monolayer contributions to the Gilbert damping in Co thin-films with different levels of local Cr doping in the upper and lower monolayers of the films. Note the calculations are for Co films capped with four monolayers (21–24) of Cu, which are not shown.

microstructural changes that can occur within the Co-Cr system at higher Cr content.²⁰

First, note that Co can exist with fcc or hcp structure and it has been shown that in thin-films the first nanometers of growth are fcc before the structure transitions to hcp, with a previous study showing that 5 nm Co sputtered films have a mix of fcc and hcp components.³⁰ With the addition of Cr up to 30%, both the fcc and hcp structures are stable,^{20,32} and for fcc Co, the lattice parameter increases by only $\sim 0.5\%$,²⁰ suggesting that the theoretical structure is a reasonable representation of the thin-film system up to at least 30% Cr doping. Nonetheless, it may be possible that the ultra-thin Cr-doped Co layer on the bottom has a different impact on the microstructure and the effective damping than the structural information above suggests due to modification of the microstructure that affects subsequent film growth. Beyond $\sim 35\%$ Cr, the initial layers of the film microstructure may become more complex, with the emergence of a tetragonal Co-Cr phase and a bcc Cr-based phase that stabilizes and dominates at higher concentrations.²⁰ These structural changes beyond 30%–40% Cr may affect the subsequent Co film growth and therefore may not provide a reasonable basis for the theoretical model, which may explain the divergence between the experimental and theoretical damping values in Fig. 4 at higher Cr doping. Previous studies have also reported changes associated with the restructuring of the energy spectrum

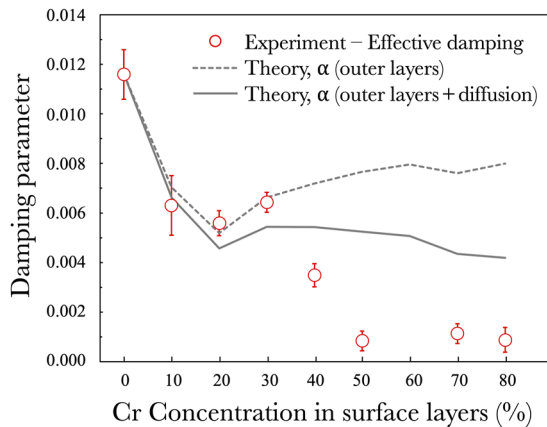


FIG. 4. Comparison of the experimental and theoretical values for damping in Co thin-films with Cr doping localized to the upper and lower monolayers of the films as a function of Cr concentration.

of Co and the emergence of resonant impurity states at 35% Cr concentration.^{33,34} This may also explain the rapid increasing of the inhomogeneous line broadening between 30% and 40% Cr. It is also reported that a long-wave absorption band can be found for Co-Cr solid solutions starting at 35% Cr that creates a new energy band above the Fermi energy level leading to interband electronic transitions.³⁴

In conclusion, it has been shown that low effective damping can be achieved in transition metal ferromagnetic thin-films by tuning the electronic structure of the upper and lower few monolayers of the thin-film by doping with another transition metal. Experiments based on Co where a few of the monolayers at the top and bottom surfaces were doped with Cr demonstrates more than an order of magnitude reduction in the effective damping from 10^{-2} to 10^{-3} . This low damping is partly explained theoretically in terms of a reduction in the contributions to the damping from the outermost atomic layers of the films achieved by doping of these outer regions of the film, while further reductions of the damping are associated with doping-induced structural changes beyond 35% Cr. This provides a novel synthetic route to develop low damping in ferromagnetic thin-films, which is applicable to the transition metal ferromagnets and their alloys.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

S. Azzawi: Conceptualization (lead); Data curation (equal); Formal analysis (lead); Investigation (equal); Methodology (equal); Visualization (lead); Writing – original draft (equal); Writing – review & editing (supporting). **A. Umerski:** Conceptualization (equal); Formal analysis (lead); Methodology (equal); Software (lead); Visualization (equal); Writing – original draft (equal); Writing – review & editing (supporting). **L. C. Sampaio:** Investigation (equal); Methodology (equal); Writing – original draft (supporting); Writing – review & editing (supporting). **S. A. Bunyaev:** Formal analysis (equal); Investigation (equal); Methodology (equal); Supervision (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **G. N. Kakazei:** Formal analysis (equal); Investigation (equal); Methodology (equal); Supervision (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **D. Atkinson:** Conceptualization (lead); Funding acquisition (equal); Investigation (lead); Methodology (equal); Project administration (lead); Supervision (lead); Writing – original draft (lead); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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