Magnetic damping in epitaxial Fe alloyed with low-atomic-number elements

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To develop low-moment, low-damping metallic ferromagnets for power-efficient spintronic devices, it is crucial to understand how magnetic relaxation is impacted by the addition of nonmagnetic elements. Here, we investigate magnetic relaxation (e.g., effective and intrinsic Gilbert damping) in epitaxial Fe films alloyed with light nonmagnetic elements of V and Al. By comparing Fe\textsubscript{100-x}V\textsubscript{x} and Fe\textsubscript{100-x}Al\textsubscript{x}, we determine whether magnetic relaxation is lowered through the reduction of the average atomic number (i.e., atomic spin-orbit coupling) or the reduction of the density of states at the Fermi level $D(E_F)$. We observe that FeV alloys exhibit lower damping compared to pure Fe, whereas damping in FeAl alloys increases with increasing Al content. Our experimental results combined with density functional theory calculations indicate that reducing $D(E_F)$, rather than the average atomic number, has a more significant impact in lowering damping in Fe alloyed with light elements.

The relaxation of magnetization dynamics (e.g., via Gilbert damping) plays important roles in many spintronic applications, including those based on magnetic switching\textsuperscript{1,2}, domain wall motion\textsuperscript{3,4}, spin wave propagation\textsuperscript{5,6}, and superfluid-like spin transport\textsuperscript{7,8}. For devices driven by spin-torque precessional dynamics\textsuperscript{1,9,10}, magnetic media with low damping and low saturation magnetization are desirable for reducing the critical current density. While some electrically insulating magnetic oxides have been considered for certain applications\textsuperscript{5,11,12}, it is essential to engineer low-damping, low-moment metallic ferromagnets for robust electrical readout with giant magnetoresistance and tunnel magnetoresistance. Prior experiments have pointed to two main contributions to intrinsic Gilbert damping in ferromagnetic metals: (1) the spin-orbit coupling strength $\xi$, which governs spin relaxation via orbital and electronic degrees of
freedom\textsuperscript{13,14}, and (2) the density of states at the Fermi level $D(E_F)$, which governs the available states for spin-polarized electrons to scatter into\textsuperscript{15,16}. 

A promising approach towards low-damping, low-moment ferromagnetic metals is to introduce nonmagnetic elements into Fe, the elemental ferromagnet with the lowest intrinsic Gilbert damping\textsuperscript{17,18}. Motivated by the premise of lowering damping through a reduced average atomic number $<Z>$ and presumably $\xi$, prior experiments have explored Fe thin films alloyed with V\textsuperscript{14,19,20}, Si\textsuperscript{21}, and Al\textsuperscript{22}. Some studies on such reduced-$Z$ Fe alloys report lower effective Gilbert damping parameters\textsuperscript{19–21} or relaxation rates\textsuperscript{14}. However, the experimentally reported damping parameters for these alloys\textsuperscript{19,20,22} are often a factor of $>2$ higher than the theoretically predicted intrinsic Gilbert damping parameter of $\approx 0.002$ in Fe\textsuperscript{23}. Furthermore, the damping parameter is often simply a quantity proportional to the linear slope of ferromagnetic resonance (FMR) linewidth versus frequency. Instead, a key quantity that captures the total magnetic relaxation at a certain excitation frequency or bias field is the FMR linewidth itself, which is governed by both intrinsic Gilbert damping and extrinsic damping due to defects and inhomogeneity. Prior experiments have in fact shown that FMR linewidths are essentially unchanged\textsuperscript{22} or increase\textsuperscript{14,19} with the introduction of low-$Z$ elements. For practical applications, it therefore remains an open challenge to realize magnetically diluted binary Fe alloys with low damping and narrow FMR linewidths. There also remains a fundamental question as to whether reduced $\xi$ or reduced $D(E_F)$ plays a more crucial role in lower damping in reduced-$Z$ Fe alloys.

Here, we investigate how magnetic relaxation is impacted by alloying epitaxial Fe with the low-$Z$ elements of V and Al. Ferromagnetic FeV and FeAl alloys possess bcc structures and constitute excellent model systems. Since Al ($Z = 13$) is a much lighter metal than V ($Z = 23$), we might expect lower magnetic relaxation in FeAl than FeV, if the smaller $<Z>$ lowers intrinsic
Gilbert damping via reduced $\zeta$. Instead, we find a significant decrease in magnetic relaxation by alloying Fe with V – e.g., a factor of $\approx 2$ smaller intrinsic Gilbert damping in FeV alloys compared to Fe – whereas damping in FeAl alloys increases monotonically with Al content. These experimental results combined with our density functional theory calculations suggest that $D(E_F)$, rather than $<Z>$, predominantly governs magnetic relaxation in binary Fe alloys containing low-Z elements. Our findings serve as an avenue for reducing damping for applications in energy-efficient spintronic devices.

Epitaxial Fe$_{100-x}$V$_x$ and Fe$_{100-x}$Al$_x$ thin films were grown using dc magnetron sputtering on (001)-oriented MgO substrates. Prior to deposition, the substrates were annealed at 600 °C for 2 hours. The base pressure prior to deposition was $< 5\times 10^{-8}$ Torr, and all films were grown with an Ar pressure of 3 mTorr. Fe and V (Al) 2” targets were dc co-sputtered to deposit Fe$_{100-x}$V$_x$ (Fe$_{100-x}$Al$_x$) films at a substrate temperature of 200 °C. By adjusting the deposition power, we tuned the deposition rate of each material (calibrated by X-ray reflectivity) to achieve the desired atomic percentage $x$ of V (Al). All FeV and FeAl films had a thickness of 25 nm, which is well above the thickness regime where interfacial effects dominate. The FeV (FeAl) films were capped with 3-nm-thick V (Al) deposited at room temperature to protect against oxidation, yielding a film structure of MgO/Fe$_{100-x}$V$_x$(25nm)/V(3nm) or MgO/Fe$_{100-x}$Al$_x$(25nm)/Al(3nm).

We confirmed the epitaxial bcc structure of our thin films using high resolution X-ray diffraction. 2θ-ω scans show only the (002) peak of the film and the (002) and (004) peaks of the substrate, as shown in Fig. 1a. Rocking curve scans of the film peaks show similar full-width-at-half-maximum values of $\approx 1.3^\circ$ irrespective of composition. The epitaxial relation between bcc Fe and MgO is well known: the bcc film crystal is rotated 45° with respect to the substrate crystal, such that the [100] axis of the film lies parallel to the [110] axis of the substrate. The
absence of the (001) film peak indicates that our epitaxial FeV and FeAl films are solid solutions rather than B2-ordered compounds\(^{29}\).

To characterize magnetic relaxation in these epitaxial FeV and FeAl films, FMR measurements (details in Supplementary Material) were performed at room temperature with the external field \(H\) applied along the in-plane [100] and [110] axes of the films. Here, unless otherwise stated, we show results for \(H \parallel [110]\) of the film. Resonance spectra for Fe, Fe\(_{80}\)V\(_{20}\), and Fe\(_{80}\)Al\(_{20}\) are shown in Fig. 1b, where we compare the peak-to-peak linewidths at a microwave excitation frequency of 20 GHz. We see that the linewidth for Fe\(_{80}\)V\(_{20}\) shows a \(\simeq 25\%\) reduction compared to Fe. We further note that the linewidth for the Fe\(_{80}\)V\(_{20}\) sample here is a factor of \(\simeq 2\) narrower than that in previously reported FeV\(^{14}\). In contrast, Fe\(_{80}\)Al\(_{20}\) shows an enhancement in linewidth over Fe, which is contrary to the expectation of lower magnetic relaxation with a lower average atomic number.

To gain further insight into the magnetic relaxation of our samples, the in-plane FMR linewidth \(\Delta H_{pp}^{IP}\) was measured over a wide range of frequencies \(f\) (Fig. 2a). Specifically, from the linear relation\(^{30}\)

\[
\Delta H_{pp}^{IP} = \Delta H_0^{IP} + \frac{h}{g\mu_B \mu_0 \sqrt{3}} \alpha_{meas}^{IP} f, \quad (1)
\]

we extract the measured Gilbert damping parameter \(\alpha_{meas}^{IP}\) and the zero-frequency linewidth \(\Delta H_0^{IP}\). In Eq. (1), \(h\) is the Planck constant, \(\mu_B\) is the Bohr magneton, \(\mu_0\) is the permeability of free space, and \(g\) is the \(g\)-factor obtained from the frequency dependence of the resonance field (see Supplementary Material). The fitting with Eq. (1) was carried out for \(f \geq 10\) GHz, i.e., sufficiently large \(H\) to saturate the films. As is evident from the results in Fig. 2a, Fe\(_{80}\)V\(_{20}\) has lower linewidths across all frequencies and a slightly lower slope, i.e., \(\alpha_{meas}^{IP}\), corresponding to a
reduction in total magnetic relaxation compared to Fe. On the other hand, Fe$_{80}$Al$_{20}$ shows higher linewidths and higher slope.

We obtain the effective in-plane Gilbert damping parameter, $\alpha_{eff}^{IP} = \alpha_{meas}^{IP} - \alpha_{eddy}$, by accounting for the small eddy current contribution $\alpha_{eddy}$ to the measured damping (Supplementary Material). As shown in Fig. 2b, $\alpha_{eff}^{IP}$ remains either invariant or slightly decreases in Fe$_{100-x}$V$_x$ up to $x = 25$, whereas we observe a monotonic enhancement of $\alpha_{eff}^{IP}$ with Al content. These results corroborate lower (higher) damping in FeV (FeAl) and suggest a factor other than the average atomic number governing magnetic relaxation in these alloys.

Besides the effective Gilbert damping parameter in Eq. (1), the zero-frequency linewidth $\Delta H_0^{IP}$ – typically attributed to magnetic inhomogeneity – often accounts for a significant fraction of total magnetic relaxation (FMR linewidths). For our samples, $\mu_0 \Delta H_0^{IP}$ is below $\approx 1$ mT (see Fig. 2b), which suggests higher film quality for our FeV samples than previously reported$^{14}$. For example, Fe$_{73}$V$_{27}$ in Scheck et al. exhibits $\mu_0 \Delta H_0^{IP} \approx 2.8$ mT, whereas Fe$_{75}$V$_{25}$ in our study exhibits $\mu_0 \Delta H_0^{IP} \approx 0.8$ mT. Although $\alpha_{eff}^{IP}$ is comparable between Scheck et al. and our study, the small $\Delta H_0^{IP}$ leads to much narrower linewidths. We speculate that the annealing of the MgO substrate prior to film deposition – a common practice for molecular beam epitaxy – facilitates high-quality epitaxial film growth and hence small $\Delta H_0^{IP}$ even by sputtering.

In the above discussions, we have implicitly attributed $\alpha_{eff}^{IP}$ to intrinsic Gilbert damping. However, $\alpha_{eff}^{IP}$ may include a contribution from two-magnon scattering due to defects$^{31}$. To test for this possibility, we performed broadband FMR with the film magnetized out-of-plane, which is the configuration that suppresses two-magnon scattering$^{25,32-34}$. For the measurements in this study, the fields were $> 4$ T, i.e. sufficient to saturate the film out-of-plane. Similar to the in-
plane data, we fit the frequency dependence of the out-of-plane FMR linewidth $\Delta H_{pp}^{OP}$ with

$$\Delta H_{pp}^{OP} = \Delta H_0^{OP} + \frac{h}{4g\mu_B\mu_0\sqrt{3}}\alpha_{meas}^{OP} f$$ (Fig. 2c). We note that the zero-frequency linewidth for the out-of-plane configuration $\Delta H_0^{OP}$ (Fig. 2c,d) is systematically greater than that for the in-plane configuration $\Delta H_0^{IP}$ (Fig. 2a,b); out-of-plane FMR measurements appear to be highly sensitive to magnetic inhomogeneity, although the exact mechanism behind this remains to be understood. Nevertheless, the absence of two-magnon scattering in out-of-plane FMR allows us to quantify the intrinsic Gilbert damping parameter, $\alpha_{int} = \alpha_{meas}^{OP} - \alpha_{eddy}$, by again subtracting the eddy current contribution.

From the compositional dependence of $\alpha_{int}$ as summarized in Fig. 2d, a clear reduction in intrinsic Gilbert damping is evidenced with V alloying. The observed minimum of $\alpha_{int} \approx 0.001$ at $x \approx 25-30$ is approximately half of the lowest Gilbert damping parameter previously reported for FeV. It is also comparable to the ultralow $\alpha_{int}$ of CoFe and Heusler alloys. The reduced intrinsic damping by alloying Fe with V is qualitatively consistent with the trend computed by Mankovksy et al. Our experimental finding confirms that FeV is an intrinsically ultralow-damping ferromagnet that possesses a smaller saturation magnetization than Fe.

For both FeV and FeAl alloys, $\alpha_{int}$ derived from out-of-plane FMR (Fig. 2d) is consistently lower than $\alpha_{eff}^{IP}$ derived from in-plane FMR (Fig. 2b). The discrepancy between $\alpha_{int}$ and $\alpha_{eff}^{IP}$ implies a two-magnon scattering contribution to magnetic relaxation in the in-plane configuration (Fig. 2b). For many applications including spin-torque oscillators and magnonic devices, it is crucial to minimize magnetic relaxation in in-plane magnetized thin films.

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1 We were unable to carry out out-of-plane FMR measurements for FeV with $x = 20$ (Fig. 2(c,d)) as the sample had been severely damaged during transit.
films. While in-plane magnetic relaxation is already low for the FeV alloys shown here, the low intrinsic Gilbert damping ($\alpha_{int} \approx 0.001$) points to the possibility of even lower relaxation by minimizing two-magnon scattering and inhomogeneous linewidth broadening. Such ultralow magnetic relaxation in FeV alloy thin films may be achieved by optimizing structural properties through growth conditions$^{27}$ or seed layer engineering$^{38}$.

The results presented so far reveal that magnetic relaxation is reduced by alloying Fe with V, whereas it is increased by alloying Fe with Al. By contrast, FeV and FeAl alloys exhibit similar compositional dependence of the effective magnetization $M_{eff}$ (here, equivalent to saturation magnetization), magnetocrystalline anisotropy field $H_k$, and the $g$-factor $g$ – all of which are quantified by fitting the frequency dependence of resonance field (Supplementary Material). As shown in Fig. 3a, there is a systematic reduction in $M_{eff}$ with increasing concentration of V and Al. We also note in Fig. 3b a gradual reduction in magnitude of the in-plane cubic anisotropy. Both of these trends are expected as magnetic Fe atoms are replaced with nonmagnetic atoms of V and Al.

The $g$-factor $g = 2 - \mu_L/\mu_S$ is related to the orbital moment $\mu_L$ and spin moment $\mu_S$; the deviation from the spin-only value of $g = 2.00$ provides insight into the strength of spin-orbit coupling $\xi$. As seen in Fig. 3c, $g$ increases by 1-2% with both V and Al alloying, which suggests that $\xi$ increases slightly with the addition of these low-Z elements. This finding verifies that $\langle Z \rangle$ is not necessarily a good predictor of $\xi$ in a solid. Moreover, the higher $g$ for FeV is inconsistent with the scenario for lower damping from reduced spin-orbit coupling. Thus, spin-orbit coupling alone cannot explain the observed behavior of Gilbert damping in Fe alloyed with low-Z elements.
We now examine whether the lower (higher) damping in FeV (FeAl) compared to Fe can be accounted for by the density of states at the Fermi level, $D(E_F)$. Utilizing the Quantum ESPRESSO package to perform density functional theory calculations (details in Supplementary Material), we calculated the density of states for Fe, Fe$_{81.25}$V$_{18.75}$, and Fe$_{81.25}$Al$_{18.75}$. For each of the binary alloys, we computed 6 distinct atomic configurations (each indicated by a band in Fig. 4) in a 2x2x2 supercell. The total density of states $D(E_F)$ is the sum of the states for the spin-up and spin-down bands. As summarized in Fig. 4 and Table I, FeV has a smaller $D(E_F)$ than Fe, whereas FeAl has a larger $D(E_F)$. These calculation results confirm a smaller (larger) availability of states for spin-polarized electrons to scatter into in FeV (FeAl), qualitatively consistent with the lower (higher) intrinsic Gilbert damping in FeV (FeAl). We thus confirm that the difference in $D(E_F)$ is a plausible governing factor for damping in FeV and FeAl alloys.

In summary, we have experimentally investigated magnetic relaxation in epitaxial Fe$_{100-x}$V$_x$ and Fe$_{100-x}$Al$_x$ thin films. We observe a reduction in intrinsic Gilbert damping to $\alpha_{int} \approx 0.001$ in FeV alloy films, whereas an increase in Gilbert damping is observed with Al content. These results cannot be explained by the change in spin-orbit coupling through alloying. Instead, we conclude that the density of states at the Fermi level plays a larger role in determining the magnitude of damping in Fe alloyed with lighter elements. Our work also confirms FeV alloys as promising ultralow-damping, low-moment materials that could be incorporated into future spintronic devices.
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The data that support the findings of this study are available from the corresponding author upon reasonable request.

|                  | Number of Spin-Up States (eV$^{-1}$) at $E_F$ | Number of Spin-Down States (eV$^{-1}$) at $E_F$ |
|------------------|---------------------------------------------|-----------------------------------------------|
| Fe               | 10.90                                       | 3.44                                          |
| Fe$_{81.25}$V$_{18.75}$ | 6.28 ± 1.80                               | 4.61 ± 0.43                                  |
| Fe$_{81.25}$Al$_{18.75}$ | 6.81 ± 1.58                                | 10.20 ± 3.03                                 |

**Table 1:** Total number of spin-up and spin-down states at $E_F$. For Fe$_{81.25}$V$_{18.75}$ and Fe$_{81.25}$Al$_{18.75}$, the average and standard deviation of values for the 6 distinct atomic configurations are shown.
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Figure 1: (a) 2θ-ω X-ray diffraction scans showing (002) and (004) substrate and (002) film peaks for bcc Fe, Fe$_{80}$V$_{20}$, and Fe$_{80}$Al$_{20}$. (b) FMR spectra at $f = 20$ GHz fitted using a Lorentzian derivative (solid line) for Fe, Fe$_{80}$V$_{20}$ and Fe$_{80}$Al$_{20}$. 
**Figure 2:** (a,b) FMR linewidth versus frequency for selected samples for field applied (a) in-plane and (c) out-of-plane. The line shows the linear fit with Eq. (1). (b,d) Alloy composition dependence of the damping parameter and zero-frequency linewidth for (b) in-plane and (d) out-of-plane FMR measurements. In (a) and (b), the data from Scheck *et al.* are of Fe$_{73}$V$_{27}$ from Ref. 14.
Figure 3: (a) Effective magnetization, (b) in-plane cubic anisotropy field, and (c) g-factor versus V and Al concentration. The solid (open) markers represent data from in-plane (out-of-plane) measurements.
Figure 4: Calculated spin-up (positive) and spin-down (negative) densities of states for (a) Fe, (b) Fe$_{81.25}$V$_{18.75}$ and (c) Fe$_{81.25}$Al$_{18.75}$. Results from the 6 distinct atomic configurations are shown in (b,c); the average densities of states at $E_F$ for Fe$_{81.25}$V$_{18.75}$ and Fe$_{81.25}$Al$_{18.75}$ are shown in Table 1.