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Anisotropic Absorption of Pure Spin Currents

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Spin transfer in magnetic multilayers offers the possibility of ultra-fast, low-power device operation. We report a study of spin pumping in spin valves, demonstrating that a strong anisotropy of spin pumping from the source layer can be induced by an angular dependence of the total Gilbert damping parameter, α , in the spin sink layer. Using lab- and synchrotron-based ferromagnetic resonance, we show that an in-plane variation of damping in a crystalline $\text{Co}_{50}\text{Fe}_{50}$ layer leads to an anisotropic α in a polycrystalline $\text{Ni}_{81}\text{Fe}_{19}$ layer. This anisotropy is suppressed above the spin diffusion length in Cr, which is found to be 8 nm, and is independent of static exchange coupling in the spin valve. These results offer a valuable insight into the transmission and absorption of spin currents, and a mechanism by which enhanced spin torques and angular control may be realized for next-generation spintronic devices.

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The generation and detection of spin currents is at the foundation of spintronics, being integral to many proposals for new memory and logic devices [1–3]. The pure spin current emitted by a ferromagnet (FM) undergoing ferromagnetic resonance (FMR) [4] is one candidate, efficiently producing torques without necessitating a charge current. The spin current can persist across a normal metal (NM), either returning to the source FM, or flowing through to a second FM, where it is absorbed and induces precession through the spin transfer torque (STT) [5]. Spin pumping can be observed by measuring increased damping (through increased FMR linewidth) due to outflow of angular momentum from the source FM [6, 7], by the inverse spin Hall effect in the sink layer [8], or by layer-specific measurements of precession or spin accumulation using x-ray magnetic circular dichroism (XMCD) [9–11].

The strength of the dynamic interaction that arises due to the pumped spin current is influenced by the transmission of spins across the FM/NM interfaces (the spin mixing conductance), and by the diffusion length of the spin current in the NM spacer layer [12, 13]. The diffusion length ranges from under a nanometer in an insulator such as MgO [8] or SrTiO₃ [14] to >100 nm in good conductors such as Ag [15]. The angular dependence of the resonance linewidth contains several contributions, including two-magnon scattering [16], mosaic broadening [17, 18], and anisotropic Gilbert damping arising due to field dragging [18].

Spin pumping can have a pronounced angular dependence, arising from the relative alignment of the two magnetic layers and the magnitude of precession [19–21]. In this case, antiparallel alignment leads to more efficient absorption of the spin current, as compensation of the pumped angular momentum drops [22]. Similarly, damp-

ing is higher when the precession of the magnetizations is out-of-phase. In trilayers with static coupling, damping can be further enhanced due to a change in the optical/acoustic character of the resonances of the two layers [23]. Furthermore, if the FM layers are simultaneously at resonance the additional damping due to the pumped spin currents can mutually cancel [24]. Stray fields from domain walls in the spin sink layer may also increase damping through an effective dipolar field [25].

In this Letter we present a study of a new mechanism for angular control of the spin current: anisotropic damping in the spin sink layer that can affect spin pumping from the source layer. Using $\text{Co}_{50}\text{Fe}_{50}/\text{Cr}/\text{Ni}_{81}\text{Fe}_{19}$ spin valves with variable Cr thicknesses, static exchange can be tuned to prefer ferromagnetic or antiferromagnetic alignment. Analysis of the resonance linewidth indicates anisotropic spin pumping in the NiFe layer, correlated with the general damping anisotropy in CoFe. This demonstrates that transmission of the spin current across the NM/FM interface into the spin sink layer is affected by damping within the FM layer. Layer resolved measurements show that the spin current pumped from the NiFe layer exerts an anisotropic torque on the CoFe layer magnetization, highlighting the potential for a new mechanism to realise angular control of spin currents.

Spin valve samples were prepared by molecular beam epitaxy (MBE) in a miniMBE system [26] on epitaxial MgO (001) substrates. The full structure is: $\text{MgO}/\text{Co}_{50}\text{Fe}_{50}(5)/\text{Cr}(t_{\text{Cr}})/\text{Ni}_{81}\text{Fe}_{19}(5)/\text{Ag}(2)$ (thicknesses in nm), with $t_{\text{Cr}} = 1, 1.5, 2, 5, 10$ nm. The sample was held at 500°C for deposition of stoichiometric $\text{Co}_{50}\text{Fe}_{50}$; epitaxial growth was observed using reflection high energy electron diffraction. The sample was then cooled to room temperature for the deposition of the Cr and $\text{Ni}_{81}\text{Fe}_{19}$ layers, to inhibit intermixing of the layers,

and ensure the Ni₈₁Fe₁₉ layer has an isotropic, polycrystalline structure.

Magnetometry measurements were carried out using a SQUID vibrating sample magnetometer (VSM). FMR measurements were performed using a vector network analyzer and octupole electromagnet. Real and imaginary components of the microwave transmission parameter, S_{12} , were measured as a function of magnetic field (strength and angle) and frequency. Plotting the resonant field as a function of frequency and bias field angle yielded anisotropy and exchange coupling parameters, with total Gilbert damping, α , extracted from the frequency dependence of the linewidth using Eq. (3).

XMCD measurements were performed on beamline I10 at the Diamond Light Source (UK) and beamline 4.0.2 at the Advanced Light Source (USA). The magnetic sample is excited by microwaves phase locked to the x-ray repetition rate, resulting in a steady precession about the effective field close to the FMR conditions. The oscillating magnetization component along the x-ray direction is probed using XMCD. Element specificity is obtained by tuning the x-ray energy to the absorption edge of the element of interest. For full details of the XFMR methodology, we refer to Ref. 27. All XFMR measurements were performed at the Ni and Co L_3 edges. Element-specific hysteresis measurements were performed at the Co, Ni, and Fe L_3 edges by sweeping the magnetic field from +100 to -100 mT and back at both x-ray helicities.

Figure 1 shows hysteresis loops measured by SQUID-VSM (left column) and element-specific XMCD-hysteresis (right column) for samples with $t_{Cr} = 1, 1.5,$ and 2 nm, in which three distinct coupling regimes can be clearly observed. For the 1 nm Cr layer [Fig. 1(a)] the strong ferromagnetic (F) interaction between the layers aligns the two magnetizations, leading to a single switching step with a coercive field of 2 mT. For $t_{Cr} = 1.5$ nm [Fig. 1(c)], however, the coupling becomes antiferromagnetic (AF), with the NiFe and CoFe layers preferring antiparallel alignment. For $t_{Cr} = 2$ nm [Fig. 1(e)] there is no coupling, and two distinct switching steps as the layers reverse independently. Precise tuning of the static exchange strength, A_{ex} , allows the contributions of static and dynamic exchange to anisotropic damping to be separated.

Figures 2(a) and 2(b) show FMR measurements of field-frequency transmission maps along the easy and hard axis of the CoFe, respectively. The anisotropic resonance showing the inflection point on the hard axis is dominated by the CoFe, whereas the more isotropic mode stems from the NiFe layer. The resonance frequency, $\omega = 2\pi f$, for in-plane magnetization is determined by the Kittel equation [28]

$$\omega = \gamma \sqrt{H_{eff}(H_{eff} + M)}, \quad (1)$$

with γ the gyromagnetic ratio, M the magnetization, and H_{eff} the effective field, composed of Zeeman, anisotropy

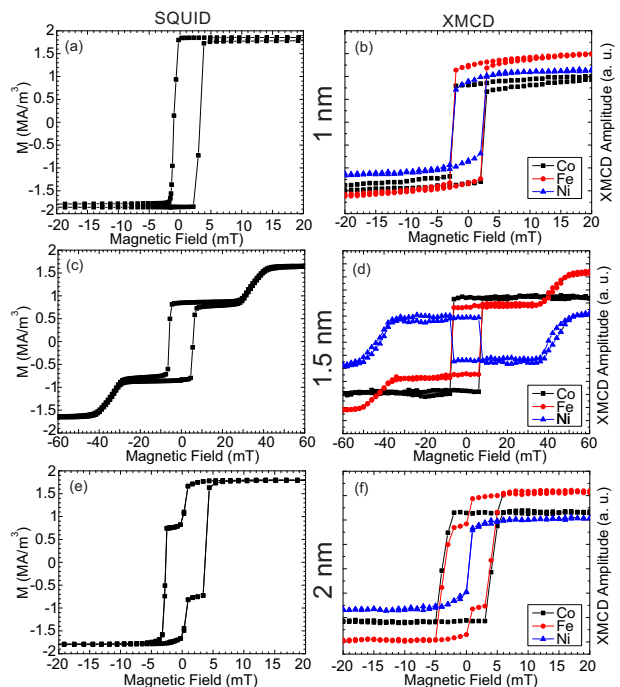


FIG. 1. (Color online) SQUID-VSM and XMCD hysteresis measurements of spin valves with $t_{Cr} = 1$ nm (a,b), 1.5 nm (c,d), and 2 nm (e,f). As the thickness of the Cr layer increases from 1 to 1.5 nm the coupling changes from F to AF before vanishing at $t_{Cr} = 2$ nm. XMCD reveals the element-specific steps in the switching, particularly the AF coupling of the Ni at low fields for $t_{Cr} = 1.5$ nm.

and exchange coupling fields. The exchange coupling field is defined as [29, 30]

$$\mu_0 H_{ex}^i = \frac{A_{ex}}{M_s^i d^i} \cos(\phi_M^i - \phi_M^j), \quad (2)$$

with A_{ex} the interlayer exchange coupling parameter (which is the same for both layers), d^i the thickness, and ϕ_M^i the alignment of the magnetization in layer i . The value of the interlayer exchange was determined by fitting the resonant field for a range of angles and fields, using M_s determined by SQUID-VSM. Example results are shown in Fig. 2(c). The CoFe resonance field varies by ± 50 mT, due to its strong magnetocrystalline anisotropy, while the NiFe resonance field has a much weaker variation of ± 5 mT, driven by a small magnetocrystalline anisotropy and exchange coupling with the CoFe layer [31]. Figure 2(d) shows A_{ex} as a function of t_{Cr} , the expected oscillatory behavior between F and AF coupling is observed [32].

The resonance linewidth, ΔH , has both intrinsic (Gilbert) and extrinsic (inhomogeneous broadening) contributions, and is given by [33]

$$\Delta H = \Delta H_0 + \frac{\alpha 4\pi f}{\gamma}, \quad (3)$$

with ΔH_0 the inhomogeneous broadening and the total

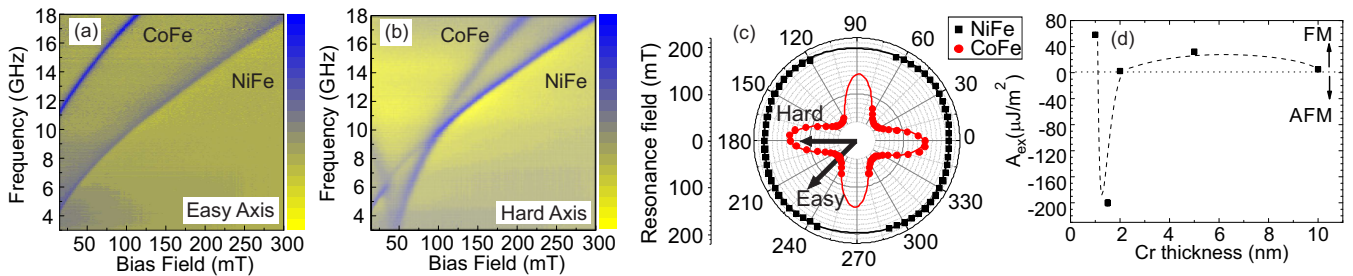


FIG. 2. (Color online) Left panels: Field vs. frequency-transmission maps, showing Kittel curves of two resonances of the $t_{\text{Cr}} = 1$ nm sample with the bias field aligned along the easy (a) and hard (b) axis of the CoFe. (c) Angular dependence of the two resonances at driving frequency 14 GHz. Solid lines are fits to the data using Eq. (1). (d) Extracted interlayer exchange coupling, A_{ex} , as a function of Cr interlayer thickness, showing F and AF coupling before being suppressed for large t_{Cr} . Error bars are comparable to the point size.

Gilbert damping $\alpha = \alpha_0 + \alpha_{\text{sp}}$. Further information on fitting can be found in Ref. [31]. The spin pumping damping α_{sp} can be inferred by plotting α as a function of interlayer thickness, as the damping associated with a bare FM, α_0 , is not affected by interlayer thickness. This is shown in Figs. 3(a) and 3(b) for the NiFe and CoFe layers, respectively. An exponential decrease in damping with increasing Cr thickness is observed, indicative of spin pumping. This arises as spins pumped from the precessing ferromagnetic layer are unable to reach the second FM, instead scattering in the spacer layer or flowing back to the spin source, where they exert an anti-damping torque. Fits to the data were performed using the procedure outlined by Kardasz and Heinrich [15], giving a spin diffusion length of 8 nm in Cr. This is somewhat less than the 13 nm reported by Du *et al.* [34] using inverse spin Hall effect measurements.

Strikingly, Fig. 3(a) shows that there is a significant anisotropy of damping in the polycrystalline NiFe layer, for $t_{\text{Cr}} \leq 2$ nm. The suppression of the anisotropy for higher values of t_{Cr} suggests that it arises from an interaction with the CoFe layer. Figure 3(b) shows that damping is also anisotropic in the CoFe layer, being significantly higher when the bias field is aligned along the magnetocrystalline hard axis. This could be indicative of inhomogeneous damping or two-magnon scattering arising from the co-ordination of defects in the crystal structure [18, 35]. The NiFe layer is examined in Fig. 3(c) in more detail, showing the angular dependence of Gilbert damping of the NiFe layer for $t_{\text{Cr}} = 1, 2,$ and 5 nm. For the thinnest Cr layer there is a strong four-fold anisotropy that correlates with the anisotropy of the CoFe, but as t_{Cr} increases this anisotropy is reduced, moving towards the isotropic damping expected of polycrystalline NiFe. The fact that the anisotropy is most pronounced for thin Cr interlayers indicates the presence of anisotropic spin pumping. In contrast to the NiFe the damping in the CoFe layer is lower, with a corresponding reduced total anisotropy that persists for all values of t_{Cr} [31].

If the static magnetizations of the two FM layers are

non-collinear, the efficiency of spin pumping is reduced due to increased backflow of spin angular momentum and partial cancellation of damping [19, 20]. This can lead to an in-plane variation of spin pumping, particularly in the case of antiparallel alignment [22, 25]. To ensure that the two magnetizations were collinear, fits to extract α were performed at fields above the anisotropy field of the CoFe. This was confirmed by calculations of the free energy derivative. Another possible source of anisotropic damping was outlined by Timopheev *et al.* [23], whereby a change in the optical/acoustic character of the modes, combined with spin pumping, causes a sharp increase in damping. To exclude this effect, fits to extract α were restricted to the region above the crossing point of the resonances; this also avoids confusion due to anti-damping arising from overlapping resonances [24]. Furthermore, the trend in anisotropy of damping as a function of t_{Cr} does not match the trend observed in the static coupling. The damping anisotropy follows a monotonic decrease with increasing t_{Cr} , while A_{ex} changes sign and oscillates over the same range. It is important to note that the damping anisotropy is present for $t_{\text{Cr}} = 2$ nm, where the static exchange coupling is close to zero [$A_{\text{ex}} = (3.3 \pm 0.6) \times 10^{-6} \text{ J m}^{-2}$].

These results therefore suggest that the spin pumping from the source layer can be effectively controlled by the in-plane damping anisotropy of the sink layer. Transmission of the spin current at the Cr/CoFe interface is then modified by the same mechanism that leads to anisotropic damping within the CoFe layer. The highest spin pumping damping occurs when the static magnetization is aligned with the direction of maximum damping in the CoFe, along the hard axis. Damping is maximized along the hard axis as most of the pumped spins are absorbed by the CoFe, which acts as a spin sink for the pure spin current ejected from the on-resonance NiFe layer. Along the easy axis, the spin current is partially reflected at the Cr/CoFe interface and returns to the NiFe, where it exerts an anti-damping torque on the precessing magnetization. This raises the possibility of a new mech-

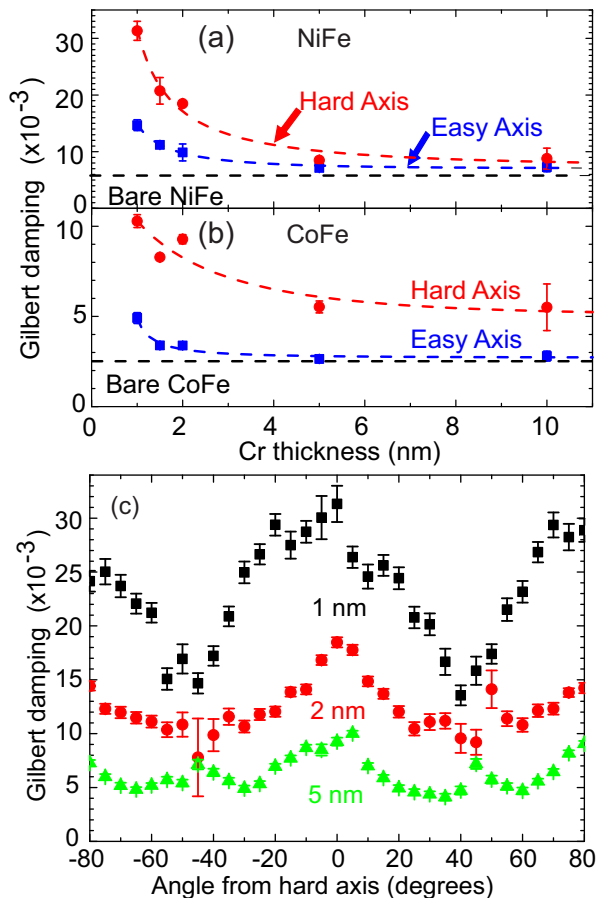


FIG. 3. (Color online) Extracted total Gilbert damping for NiFe (a) and CoFe (b) layers, measured along the easy and hard axes of the CoFe layer. The flat line indicates the damping of the bare ferromagnets, while dashed lines are fits to the data that yield a spin diffusion length of 8 nm. The observed anisotropy of damping is shown in more detail in (c), plotting the Gilbert damping of the NiFe layer as a function of bias field angle with respect to the hard axis of the CoFe layer.

anism for fine control of spin pumping through modification of sink layer damping parameters and engineering of the NM/FM interfaces.

Layer-resolved measurements of the magnetodynamics were performed with XFMR to separate the effects of static and dynamic exchange, and investigate the anisotropic spin transfer in more detail. A driving frequency of 4 GHz was used to excite precession across the NiFe resonance at ~ 14 mT, while the XMCD amplitudes at the Co and Ni L_3 edges were measured. The relative phase of microwave excitation and x-ray pulse was varied, extracting amplitude and phase of precession from plots of time-dependent magnetization [9, 27]. Results are plotted in Fig. 4, showing the phase of precession with the bias field aligned along the easy axis (a), an intermediate axis of 22° away from the easy axis (b), and along the hard axis (c). These measurements show how the anisotropic spin pumping affects the spin transfer torque

exerted on the magnetization of the CoFe layer. The expected phase shift of almost 180° is observed in the on-resonance Ni data, while Co shows significant induced precession.

XFMR results can be modeled by a linearized macrospin solution of the LLG, incorporating both static coupling and dynamic exchange through spin pumping [9, 24]:

$$-\frac{\partial \mathbf{m}_i}{\partial t} = \gamma_i \mathbf{m}_i \times (\mathbf{H}_{\text{eff}}^i + \beta_i M_{s,j}) - (\alpha_i^0 + \alpha_{ii}^{\text{sp}}) \frac{\partial \mathbf{m}_i}{\partial t} + \alpha_{ij}^{\text{sp}} \mathbf{m}_j \times \frac{\partial \mathbf{m}_j}{\partial t}, \quad (4)$$

where the subscript denotes the magnetic layer, β_i the static exchange coupling, and α_{ii}^{sp} additional damping due to spin pumping out of layer i , and α_{ij}^{sp} anti-damping due to pumping into layer i from layer j . Solving Eq. (4) yields the AC susceptibility, from which amplitude and phase of the precession were determined. This approach uniquely determines the different contributions of static and dynamic exchange, and allows increased spin pumping to be separated from increased intrinsic damping. According to this model, static coupling leads to a unipolar (absorptive) feature in the phase, while dynamic coupling leads to a bipolar (dispersive) feature, the combination of the two leads to an asymmetric step-like feature; examples are shown in Fig. 4(d). Solid lines in Figs. 4(a-c) are results from this model using parameters extracted from analysis of Kittel curves (Fig. 2) and angle-dependent damping (Fig. 3). Close agreement with the XFMR data confirms the presence of strongly anisotropic spin pumping, as opposed to increased intrinsic damping or static exchange, and shows the importance of considering both exchange mechanisms when modeling coupled magnetodynamics. As spin pumping increases the phase shift curvature increases, transitioning from static exchange-dominated unipolar, to dynamic exchange-dominated bipolar. This behavior is indicative of an increase in STT exerted on the Co moments.

In summary, we have studied the magnetization dynamics of $\text{Co}_{50}\text{Fe}_{50}/\text{Cr}(t_{\text{Cr}})/\text{Ni}_{81}\text{Fe}_{19}$ trilayers as a function of t_{Cr} . We find a significant anisotropy of spin pumping from the NiFe layer, arising from an anisotropy of Gilbert damping in the CoFe layer. This suggests that the transmission across the spacer layer is governed by the characteristics of the spin sink as well as the spin source. This anisotropy is unaffected by the strength or character of the static exchange coupling, but is suppressed as t_{Cr} exceeds the spin diffusion length in Cr, at ~ 8 nm. We attribute the observed angle-dependence to anisotropic transmission and reflection of spins at the Cr/CoFe interface. Layer-resolved measurements of induced precession in the CoFe at the NiFe resonance at 4 GHz confirm this observation, revealing the competition between static and dynamic exchange that occurs in such spin valves. These results shed new light on the

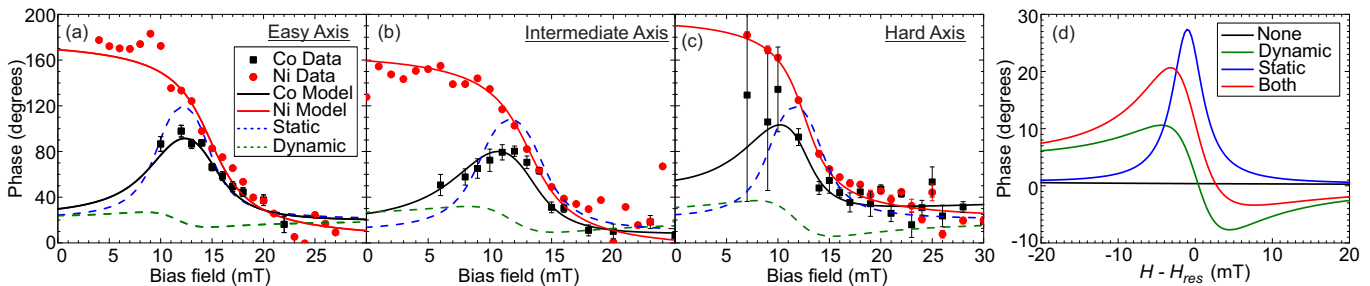


FIG. 4. (Color online) Phase of precession for Co (black squares) and Ni (red circles) moments as measured by XFMR for the $t_{Cr} = 1$ nm sample with the bias field along the easy (a), intermediate (22° away) (b), and hard (c) axes of the CoFe layer. Solid lines are model calculations using parameters extracted by fitting of Kittel curves (Fig. 2) and damping (Fig. 3). Dashed lines show the effects of dynamic coupling only (green), and static coupling only (blue). The large error bars in (c) are due to canting of magnetization reducing the projection along the beam, in turn reducing the measured XMCD signal. (d) Differing effects of static and dynamic exchange on induced precession. Note that the couplings can also interfere with each other, e.g., the increased linewidth from the dynamic interaction leads to a larger phase shift from the static interaction. The dashed lines corresponding to static-only are for a spin valve with no spin pumping, and therefore are the same for all magnetization orientations. In the case of the dynamic-only dashed lines, turning off the static coupling causes the resonance field to shift by a few mT; this effect has been removed for ease of comparison.

increasingly important topic of the anisotropic generation and detection of spin currents. Furthermore, they suggest the possibility of further control of spin pumping through magnetization alignment, as well as suggesting new concepts to manipulate spin pumping, through modification of the damping mechanisms in the spin sink layer.

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