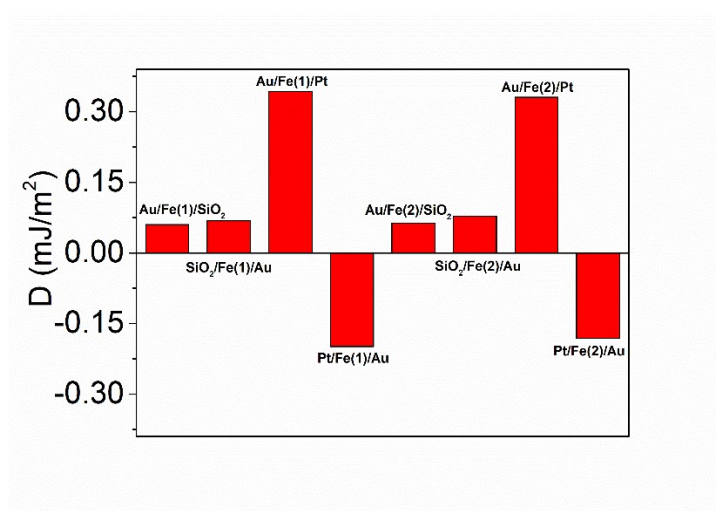


## Supplementary Note 1: The effect of thickness of Fe on the chirality of i-DMI

In this section, we demonstrated the influence of the thickness of Fe on the chirality of i-DMI in Au/Fe structures. As shown in Supplementary Figure 1, the results were quite similar as seen in Figure 2h, and the chirality was the same no matter when Au is on the top or at the bottom surface of Fe. Clearly, the chirality of i-DMI changed to the right-handed at the Au/Fe interface for all samples. Moreover, as the thickness of Fe decreased from 3 nm down to 1 nm, the anisotropy energy increased from  $3 \times 10^6$  erg/cc to  $5 \times 10^6$  erg/cc (see table 1). However, as illustrated in Supplementary Figure 1, the strength of i-DMI remains almost unchanged. These results further confirmed the negligible influence of anisotropy on i-DMI in our samples.

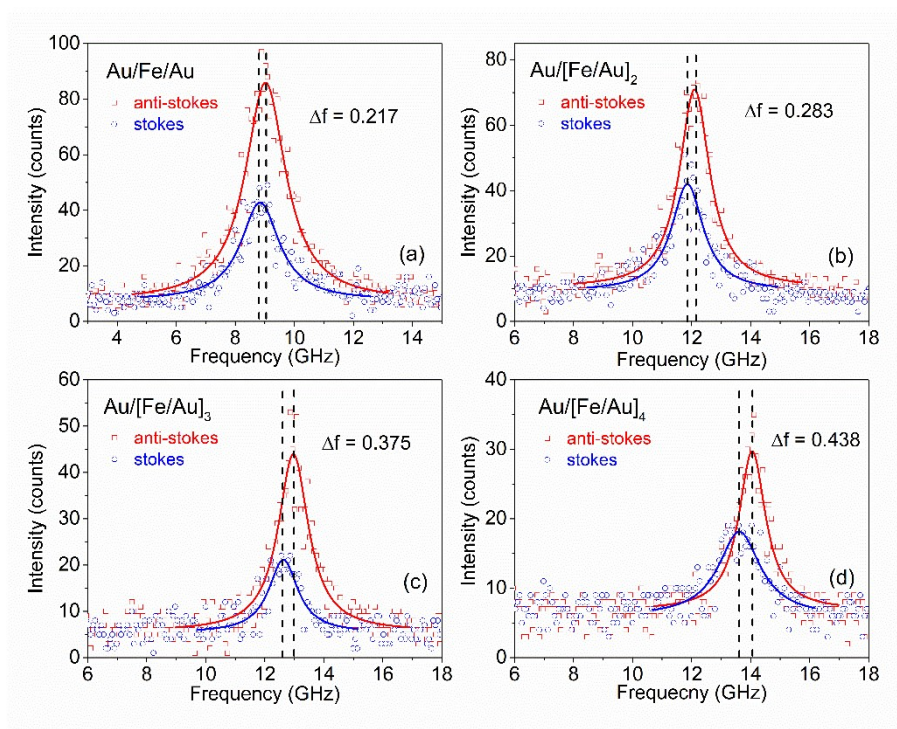


**Supplementary Figure 1** | The magnitude of i-DMI extracted from BLS measurements for the different samples.

**Supplementary Note 2: Additional BLS spectra for Au/[Fe/Au]<sub>n</sub>**

### systems with different repetition number n.

In this section, we display the typical BLS spectra for  $\text{Au}/[\text{Fe}/\text{Au}]_n$  multilayers as shown in Supplementary Figure 2. It was obvious that the frequency shift  $\Delta f$  increased with the increasing repetition number n.

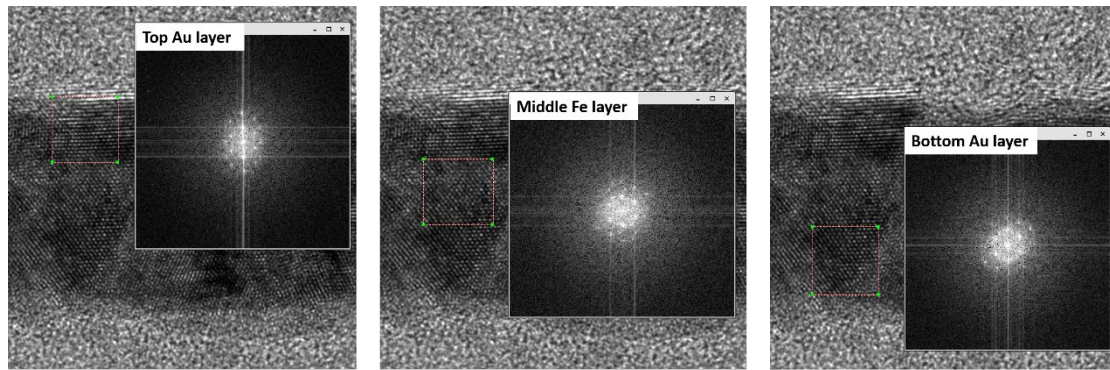


**Supplementary Figure 2** | (a)-(d) BLS spectra (open symbols) measured for the  $n = 2 - 4$  samples in  $\text{Au}/[\text{Fe}/\text{Au}]_n$  system, where the wave vector  $k$  is  $1.518 \times 10^7$  rad/m. The magnetic field is 500 Oe for  $\text{Au}/\text{Fe}/\text{Au}$  sample, and 1000 Oe for the others. The solid lines are Lorentzian fits. The shift in frequency  $\Delta f = |f_{\text{as}}| - |f_{\text{s}}|$ , where  $f_{\text{as}}$  and  $f_{\text{s}}$  correspond to frequency of anti-Stokes and Stokes peaks.

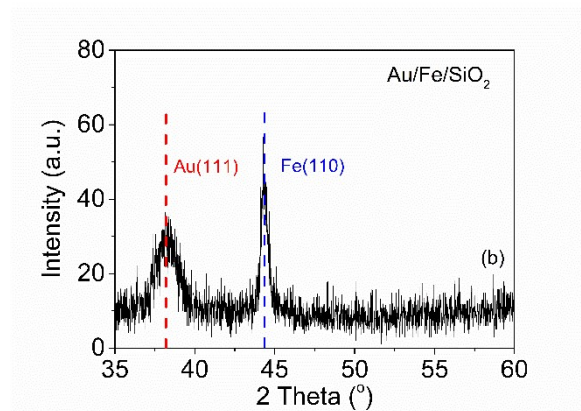
### Supplementary Note 3: The structure of Au/Fe/Au system

Here we display the Fourier transform (FFT) of HRTEM images in different regions (see Supplementary Figure 3a). The results roughly

exhibited a “quasi-epitaxial” structure. In Supplementary Figure 3b, we clearly show the preferred orientations of Fe and Au with (110) and (111) textures, respectively.



(a)



**Supplementary Figure 3** | (a) FFT images of HRTEM for top Au, middle Fe, and bottom Au layers. (b) XRD pattern of Au/Fe bilayers system.