Supplementary information

1) Calculus of microstrain, ϵ, for each $Ni_{100-x}Fe_x$

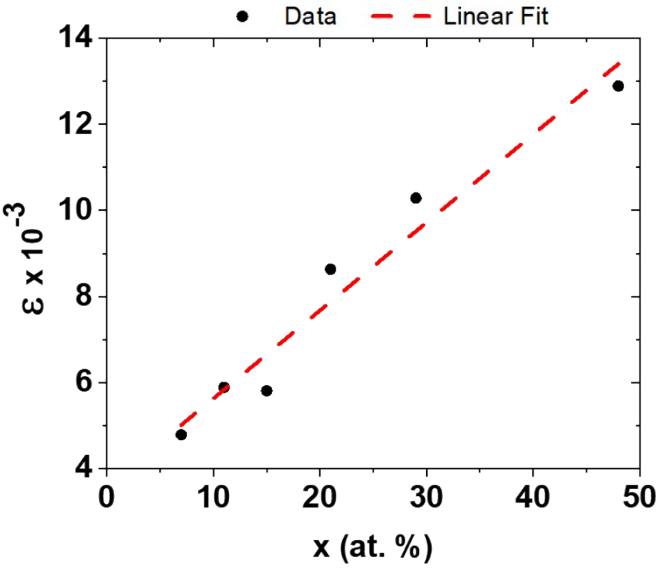
From the XRD measurements, we obtain the following data from the (111) diffraction peak:

x (at. %)	$\theta \times 10^{-2}$ (rads)	FWHM $\times 10^{-2}$ (rads)	$\epsilon \times 10^{-3}$
7	38,771	0,782	4,788
11	38,709	0,961	5,891
15	38,756	0,949	5,809
21	38,752	1,410	8,634
29	38,592	1,672	10,288
48	38,745	2,105	12,893

where θ is the center of the (111) peak, FWHM is the full width half maximum, and ϵ is the microstrain, which is calculated as:

$$\epsilon = \frac{FWHM}{4\tan\theta} \qquad (1)$$

The plot of ϵ *vs* x is shown in Supp. Fig. 1, where a linear fit is performed. The line equation obtained is $\epsilon = 2.05 \times 10^{-4} \cdot x + 3.58 \times 10^{-3}$. By using this equation, we can estimate the values of ϵ for all compositions of the $Ni_{100-x}Fe_x$ alloys, enabling us to calculate the magnetoelastic anisotropy present in the films.



Supplementary Figure 1. Microstrain as a function of the Fe at. %. Experimental data in black circles and linear fit in dashed red line.

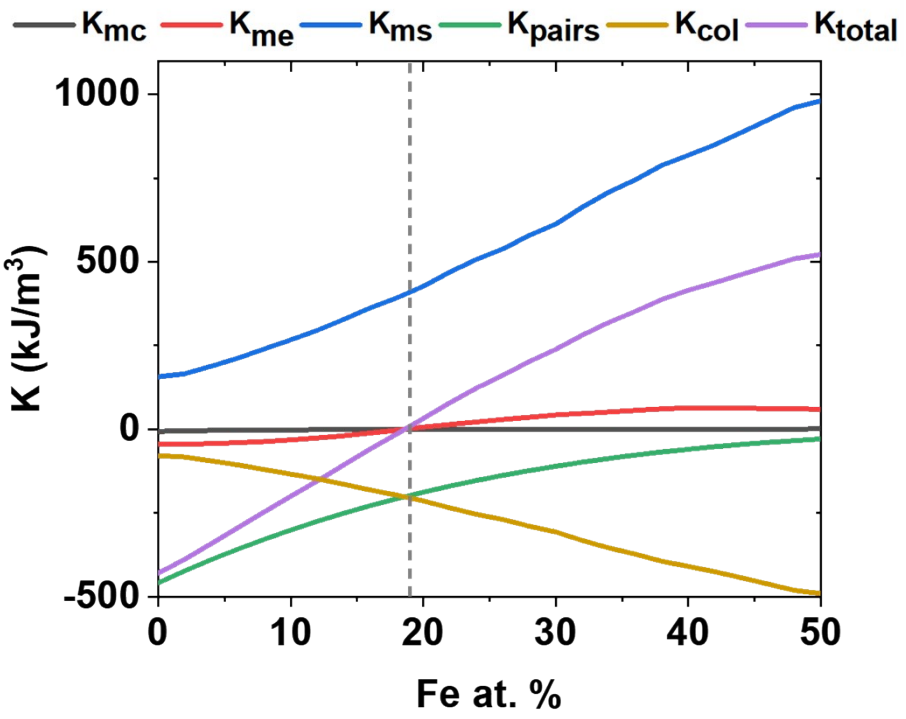
2) Effect of columnar growth on the model

By adding an anisotropy term to account for the columnar growth, K_{col}, our model yields:

$$K_{tot} = K_{mc} + K_{me} + K_{ms} + K_{pairs} + K_{col}$$

Where the other sources of magnetic anisotropy are defined in the text of the manuscript: K_{mc}: magnetocrystalline anisotropy; $K_{me} = \frac{3}{2}\lambda_p \epsilon Y$; $K_{ms} = \frac{1}{2}\mu_0 M_s^2$; $K_{pairs} = -\frac{4K^2C^4a^2}{\pi A_0}$, with $K = 3.30 \times 10^6$ J/m^3.

Considering that the contribution of the columnar growth is out-of-plane ($K_{col} < 0$) and can be modeled as a magnetostatic energy, we can define the columnar anisotropy as $K_{col} = -\frac{1}{2}NM_s^2$ being N, the demagnetization factor induced by the columnar shape. Assuming the columnar growth as a perfect cylinder $N = 0.5$, we obtain that the change of sign in total anisotropy occurs at Fe 19 at. %.



Supplementary Figure 2. Dependency of the different magnetic anisotropies with respect to Fe content (at.%). In this figure, columnar growth is considered as a source of OOP anisotropy. The dashed line at Fe 19 at.% indicates the change of sign in K_{tot}.