Supporting Information for:

Perpendicular Magnetic Anisotropy and Magneto-Optical Properties of Bi, Mn:YIG Epitaxial Films

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Table S1 The results of the compositional analysis (EDS) of the samples $(Y_{3-x}Bi_x)(Fe_{5-y}Mn_y)O_{12}$

samples	Atomic percent(%)					x_{Bi}	y_{Mn}
	O	Fe	Y	Bi	Mn		
BYG03	65.90	20.24	12.17	1.56	-	0.34	0
BMYG04	59.92	23.40	14.25	0.98	0.38	0.19	0.08
BMYG05*	59.93	24.05	13.74	1.27	0.35	0.25	0.07
BMYG06*	59.86	23.31	14.41	0.57	0.76	0.11	0.16
BMYG09	59.73	22.67	15.17	0.50	1.37	0.10	0.28

Raman spectra were used to characterize crystallinity of samples. The full width at half maxima (FWHM) of strongest peak (270cm^{-1}) of samples is basically about 17cm^{-1}. In comparison, the BMYG05 has a slightly less crystallinity.

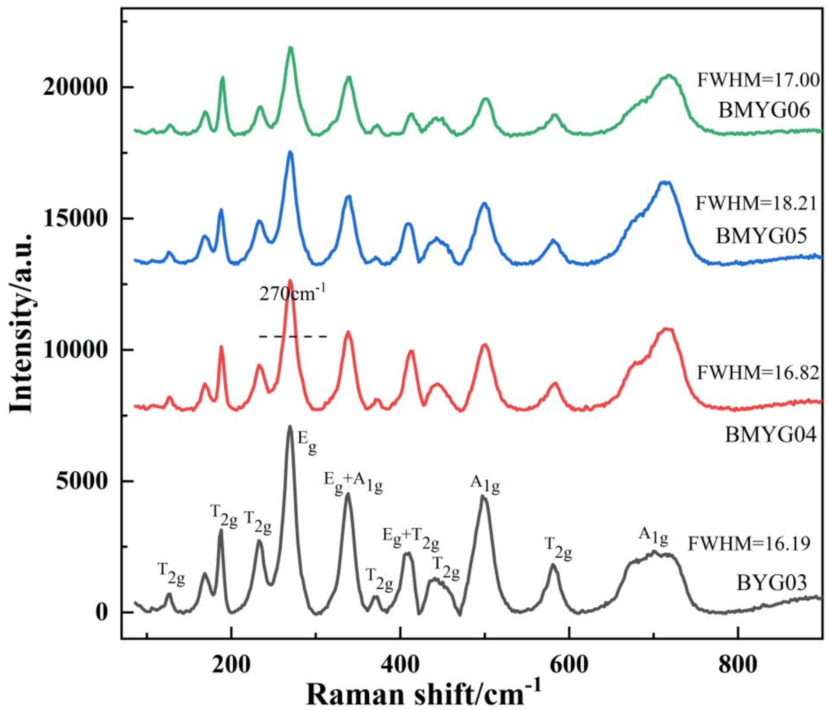

Figure S1 The Raman spectra of the samples

The calculation of the lattice constant considers both the K_{a1} peak and the K_{a2} peak. Since YIG belongs to the cubic system, the in-plane and out-of-plane lattice constants of the relaxed YIG are equal.

$$a_{f0\perp} = a_{f0//} = a_{f0} \tag{S1}$$

The difference $\Delta a_{\perp}$ between the lattice constant of the substrate and that of the strained YIG can be expressed as

$$\Delta a_{\perp} = a_s - a_{f\perp} \tag{S2}$$

Where a_s is the lattice constant of the substrate, $a_{f\perp}$ is the out-of-plane lattice constant of the strained YIG. Since the thickness (0.5mm) of the substrate is much larger than the films, we can think that the lattice constant of the substrate remains constant during the formation process of the pseudomorphic structure. The out-of-plane lattice constant of the substrate and that of the strained YIG can be calculated through the XRD patterns. The lattice constant of the relaxed YIG can be expressed as

$$a_{f0} = a_{f0\perp} = a_s - \Delta a_{\perp} \times \frac{1-\nu}{1+\nu} \tag{S3}$$

Where ν is the Poisson's ratio ($\nu = 0.286$). The out-of-plane lattice strain $\varepsilon_{\perp}$ of the strained YIG can be expressed as

$$\varepsilon_{\perp} = \frac{a_{f\perp} - a_{f0\perp}}{a_{f\perp}} \tag{S4}$$

The in-plane lattice strain $\varepsilon_{//}$ of the strained YIG can be expressed as

$$\varepsilon_{//} = -\frac{C_{11} + 2C_{12} + 4C_{44}}{2(C_{11} + 2C_{12} - 2C_{44})}\varepsilon_{\perp} \tag{S5}$$

Where the modulus of elasticity of the YIG include: $C_{11} = 2.68 \times 10^{11}\,\mathrm{N/m^2}$, $C_{12} = 1.106 \times 10^{11}\,\mathrm{N/m^2}$, $C_{44} = 0.766 \times 10^{11}\,\mathrm{N/m^2}$. The in-plane lattice constant $a_{f//}$ of the strained YIG can be expressed as

$$a_{f//} = (1 + \varepsilon_{//})a_{f0//} \tag{S6}$$

The in-plane lattice stress $\sigma_{//}$ of the strained YIG can be expressed as

$$\sigma_{//} = 6C_{44}\frac{C_{11} + 2C_{12}}{C_{11} + 2C_{12} + 4C_{44}}\varepsilon_{//} \tag{S7}$$

The magnetic anisotropy of the films is mainly composed of magnetocrystalline anisotropy, shape anisotropy, stress induced anisotropy, growth-induced magnetic anisotropy, etc. Since we cannot directly measure the magnetocrystalline anisotropy, we only discuss the change of the magnetic anisotropy of the samples, not the absolute values. Due to the large thickness of the samples (> 10 μm), the shape anisotropy (about 10^{-1} erg/cm^3) can be ignored. The actual magnetic anisotropy is obtained from the hysteresis loops in the IP and OP directions (see dashed lines in Figure 6). H_A is the transverse coordinate of the intersection of the initial magnetization curve in the IP direction and the horizontal line of saturation magnetization in the OP direction. The actual magnetic anisotropy constant can be expressed as

$$K_{\mathrm{eff}} = \frac{H_A \times M_S}{2} \tag{S8}$$

The stress-induced magnetic anisotropy constant can be expressed as

$$K_{\mathrm{indu}} = -\frac{3}{2}\sigma_{//}\lambda_{111} \tag{S9}$$

Where λ_{111} is the magnetostriction coefficient (-2.4*10^{-6}). The growth-induced magnetic anisotropy caused by magnetostriction can be expressed as

$$K_{\mathrm{magnet}} = b \times \xi \tag{S10}$$

Where b is the magnetoelastic constant, which is obtained from ref.[16] The ξ is the lattice strain, $\xi = (a_{OP} - a_{IP})/a_{IP}$. From the discussion of the pseudomorphic structures, the magnetostriction effect of sample BMYG04 is opposite. Calculations of the magnetic anisotropy are shown in the Table S1 and Figure S1.

Table S2 Calculations of the magnetic anisotropy

samples	y_{Mn}	M_S/emu*cm^{-3}	H_A/Oe	K_{eff}/erg*cm^{-3}	K_{indu}/erg*cm^{-3}	$10^4\xi$	K_{magnet}/erg*cm^{-3}
BYG03	0	0.1057	4474.4	236.425	27144	48.953	42366.49
BMYG04	0.08	0.1629	-2372.3	-193.218	33948	63.799	-38167.99
BMYG06	0.16	0.2325	3153.1	366.588	35892	65.008	21521.24
BMYG09	0.28	0.1463	3093.0	226.196	32976	59.7865	-4169.672

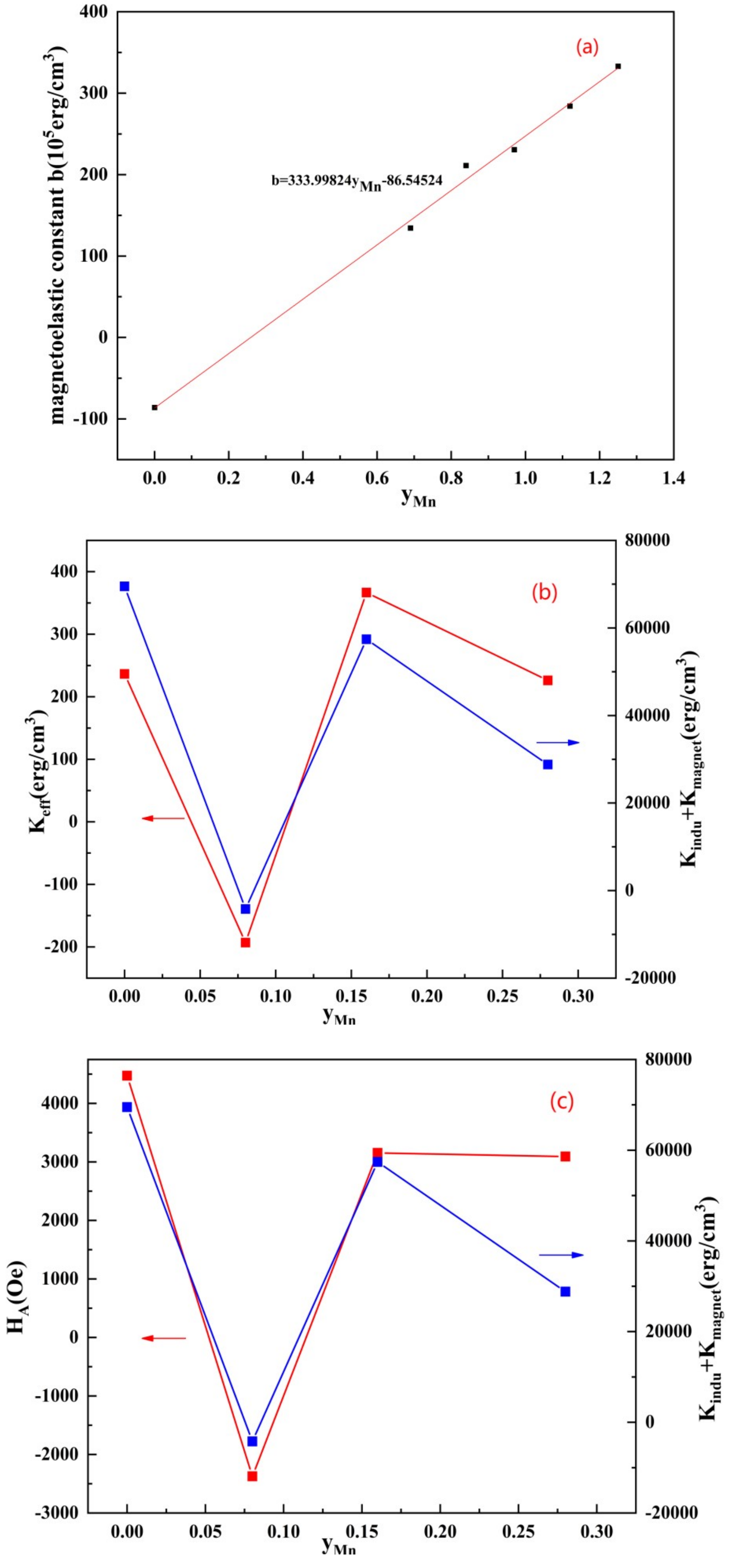

Figure S2 (a) the magnetoelastic constant against the Mn^{3+} content, (b-c) actual magnetic anisotropy and the calculated magnetic anisotropy against the Mn^{3+} content.