

Sample and Experimental Details

The original diameter of the $(\text{Mn}_{0.85}\text{Fe}_{0.15})\text{Si}$ crystal was larger than 7 mm. We changed it to a diameter slightly smaller than 7 mm to carry out the first PSI experiment (0.0, 11.7, 14.1 and 16.4 kbar). After that, we changed it to a diameter slightly smaller than 6 mm to get higher pressure for the second PSI experiment (6.0, 7.8, 19.3 and 23 kbar). The ratio of muons stopping at the pressure cell for 6.0, 7.8, 19.3 and 23 kbar are higher than those for 0.0, 11.7, 14.1 and 16.4 kbar.

Hydrostatic pressure was generated by using a piston-cylinder-type pressure cell made of non-magnetic alloy Ni-Co-Cr-Mo (MP35N). The sample space was filled with a pressure-transmitting medium of Daphne 7373. Pressure in the sample space was calibrated from the pressure dependence of superconducting temperature of lead. We used two pressure cells. The cell with $\varnothing 7$ mm and $\varnothing 6$ mm inside diameter can reach up to 19 kbar and 23 kbar, respectively.

P (kbar)	ZF	LF	WTF	Pressure cell	Sample	Place
0.0		LF			Cylindrical, slightly smaller than $\varnothing 7$ mm	TRIUMF
0.0	ZF		WTF	$\varnothing 7$ mm	Cylindrical, slightly smaller than $\varnothing 7$ mm	PSI
11.7	ZF	LF	WTF	$\varnothing 7$ mm	Cylindrical, slightly smaller than $\varnothing 7$ mm	PSI
14.1	ZF	LF		$\varnothing 7$ mm	Cylindrical, slightly smaller than $\varnothing 7$ mm	PSI
16.4	ZF	LF		$\varnothing 7$ mm	Cylindrical, slightly smaller than $\varnothing 7$ mm	PSI
6.0	ZF	LF		$\varnothing 6$ mm	Cylindrical, slightly smaller than $\varnothing 6$ mm	PSI
7.8		LF		$\varnothing 6$ mm	Cylindrical, slightly smaller than $\varnothing 6$ mm	PSI
19.3	ZF	LF		$\varnothing 6$ mm	Cylindrical, slightly smaller than $\varnothing 6$ mm	PSI
23	ZF	LF		$\varnothing 6$ mm	Cylindrical, slightly smaller than $\varnothing 6$ mm	PSI

ZF Function

$$A_{\text{sample}} \left[f_{\text{mag}} \exp(-\lambda t) \left\{ \frac{1}{3} + \frac{2}{3} (1 - (\sigma t)^\beta) \exp\left(-\frac{(\sigma t)^\beta}{\beta}\right) \right\} + (1 - f_{\text{mag}}) \exp(-\lambda t) \right] \\ + A_{\text{cell}} \exp(-(\Delta t)^\gamma) \left\{ \frac{1}{3} + \frac{2}{3} (1 - (\Delta t)^2) \exp\left(-\frac{(\Delta t)^2}{2}\right) \right\} + A_{\text{background}}$$

where

$$\Lambda = 0.060 + 1.0402 \times \exp(-4.83 \times T) [\mu s^{-1}]$$

$$\gamma = 1.26$$

$$\Delta = 0.325 [\mu s^{-1}]$$

$$A_{\text{background}} = 0.110 \times A_{\text{total}} = 0.110 \times 0.275 \approx 0.0301$$

A_{total} is fixed at 0.275. We can rewrite the function as follows:

$$A_1 \exp(-\lambda t) \left\{ \frac{1}{3} + \frac{2}{3} (1 - (\sigma t)^\beta) \exp\left(-\frac{(\sigma t)^\beta}{\beta}\right) \right\} + A_2 \exp(-\lambda t) \\ + A_{\text{cell}} \exp(-(\Delta t)^\gamma) \left\{ \frac{1}{3} + \frac{2}{3} (1 - (\Delta t)^2) \exp\left(-\frac{(\Delta t)^2}{2}\right) \right\} + A_{\text{background}}$$

where

$$A_1 = f_{\text{mag}} A_{\text{sample}}$$

$$A_2 = (1 - f_{\text{mag}}) A_{\text{sample}} = A_{\text{total}} - A_1 - A_{\text{cell}} - A_{\text{background}} = 0.245 - A_1 - A_{\text{cell}}$$

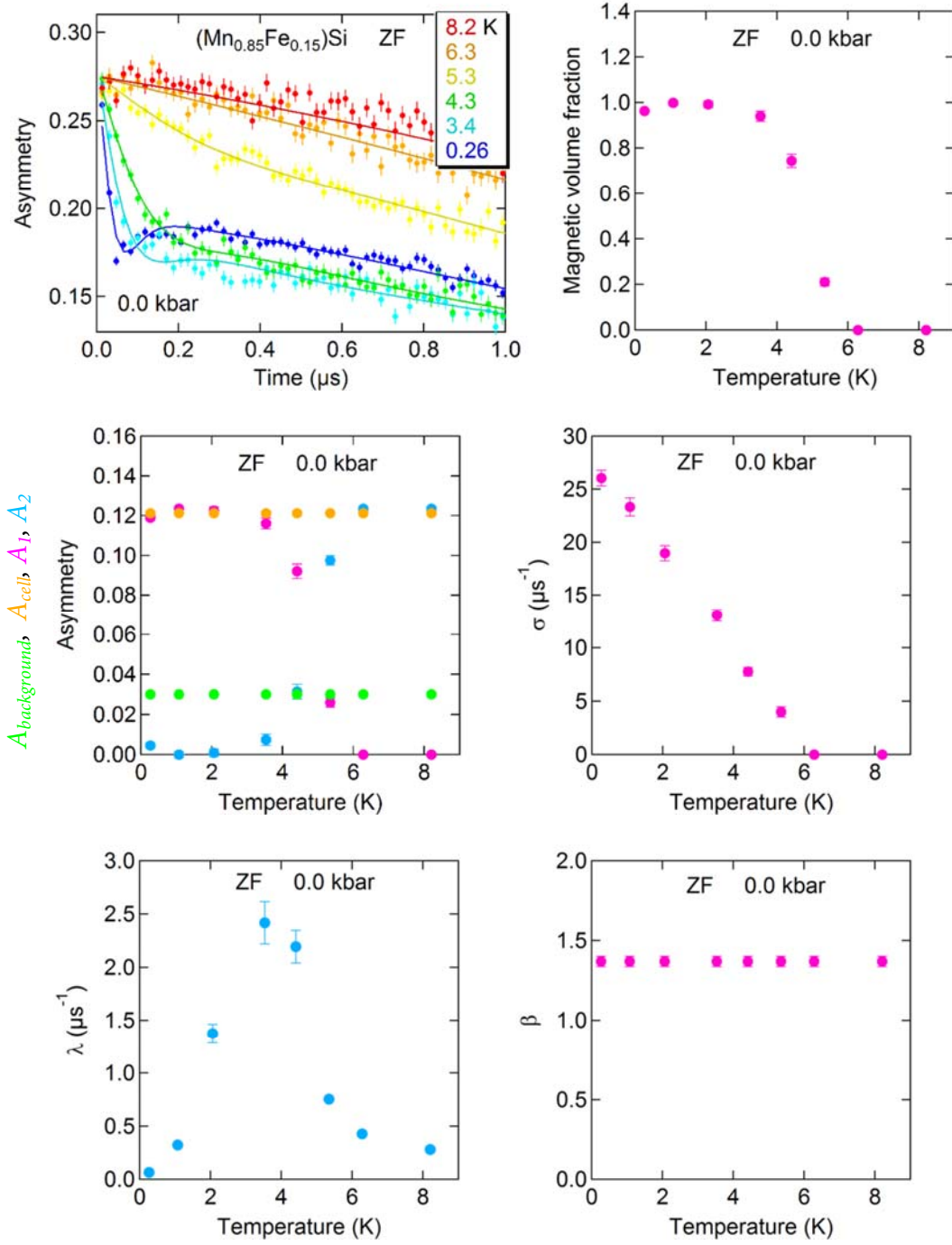
A_1 , λ and σ are a free parameter. β is common to the temperature dependence. A_{cell} is a global parameter for the 0.0 kbar data, and then A_{cell} is fixed at the value for 11.7, 14.1 and 16.4 kbar. Also, A_{cell} is a global parameter for the 6.0 kbar data, and then A_{cell} is fixed at the value for 19.3, 23 kbar. The fitting time range is 0.005-3.025 μs .

Figure Caption: Supplementary Information Figure 1.

Time spectra and extracted parameters from Zero-Field (ZF) MuSR measurements of (Mn,Fe)Si

Observed time spectra, the signal asymmetries, the dynamic relaxation rate λ , the volume fraction of the magnetically ordered regions, the static relaxation rate σ from the magnetically ordered volume, and the stretching power parameter β in the generalized Kubo-Toyabe function, obtained from the Zero-Field MuSR measurements in a single crystal of (Mn,Fe)Si with 15% Fe in (a) ambient pressure, (b) at hydrostatic pressure of 11.7 kbar, (c) 14.1 kbar, (d) 16.4 kbar, (e) 6.0 kbar, (f) 19.3 kbar. (g) shows the results obtained in an empty pressure cell without the (Mn,Fe)Si crystal.

(a) ZF 0.0 kbar



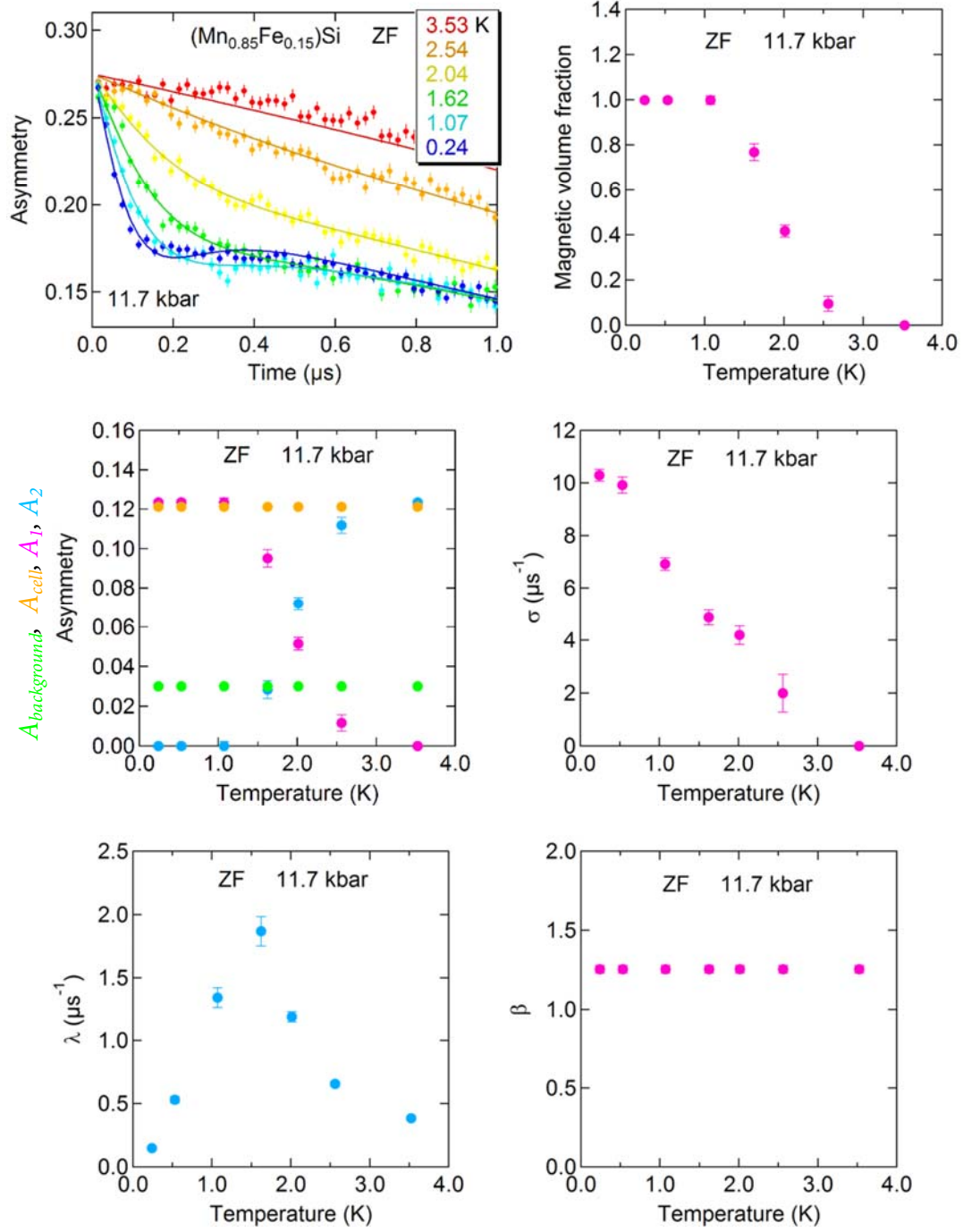
$$A_{\text{cell}} = 0.121$$

$$A_{\text{sample}} = A_{\text{total}} - A_{\text{cell}} - A_{\text{background}} = 0.124$$

$$A_{\text{sample}} : A_{\text{cell}} + A_{\text{background}} = 0.449 : 0.551$$

$$f_{\text{mag}} = \frac{A_1}{A_{\text{sample}}} = 8.09 \times A_1$$

(b) ZF 11.7 kbar



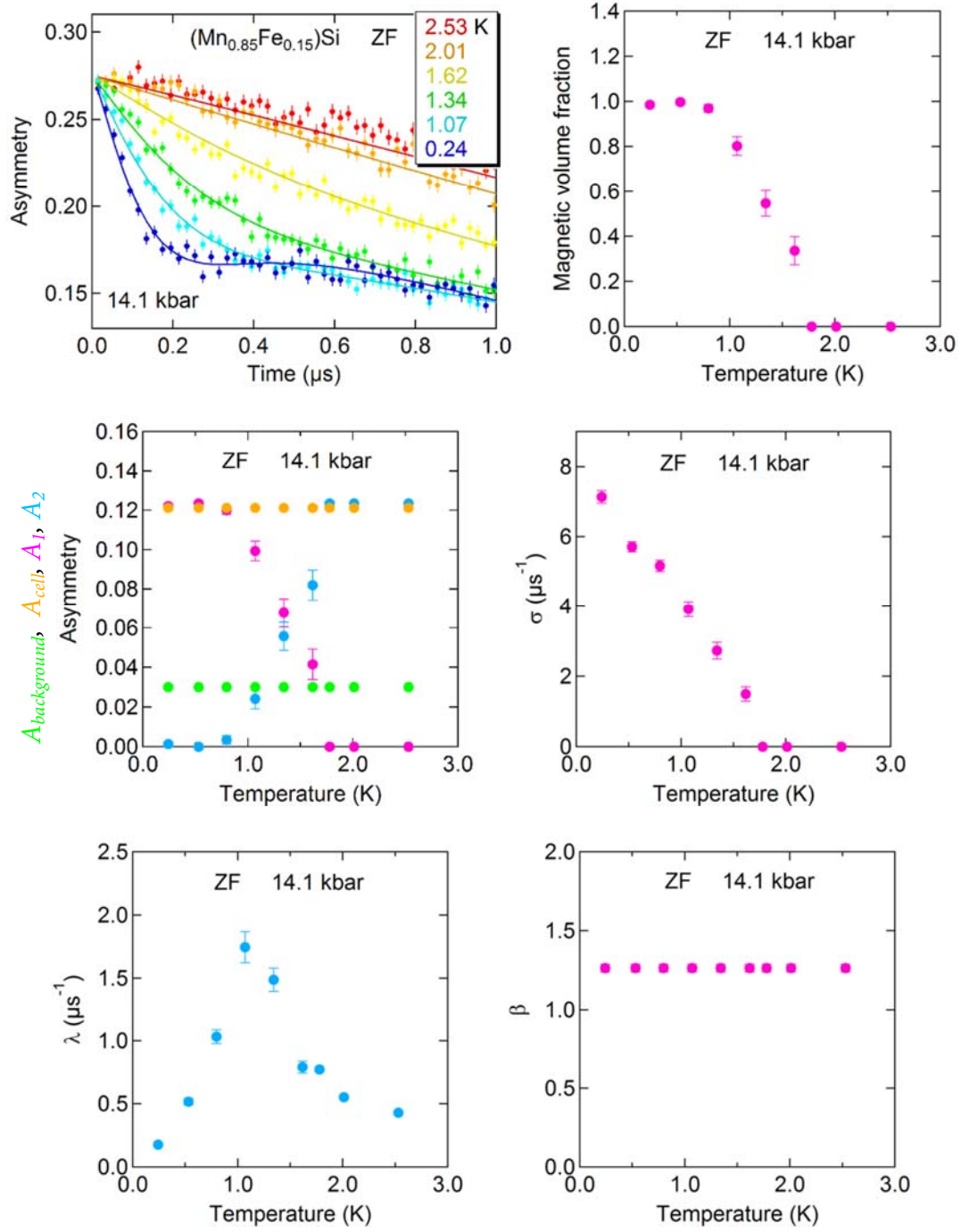
$$A_{\text{cell}} = 0.121$$

$$A_{\text{sample}} = A_{\text{total}} - A_{\text{cell}} - A_{\text{background}} = 0.124$$

$$A_{\text{sample}} : A_{\text{cell}} + A_{\text{background}} = 0.449 : 0.551$$

$$f_{\text{mag}} = \frac{A_1}{A_{\text{sample}}} = 8.09 \times A_1$$

(c) ZF 14.1 kbar



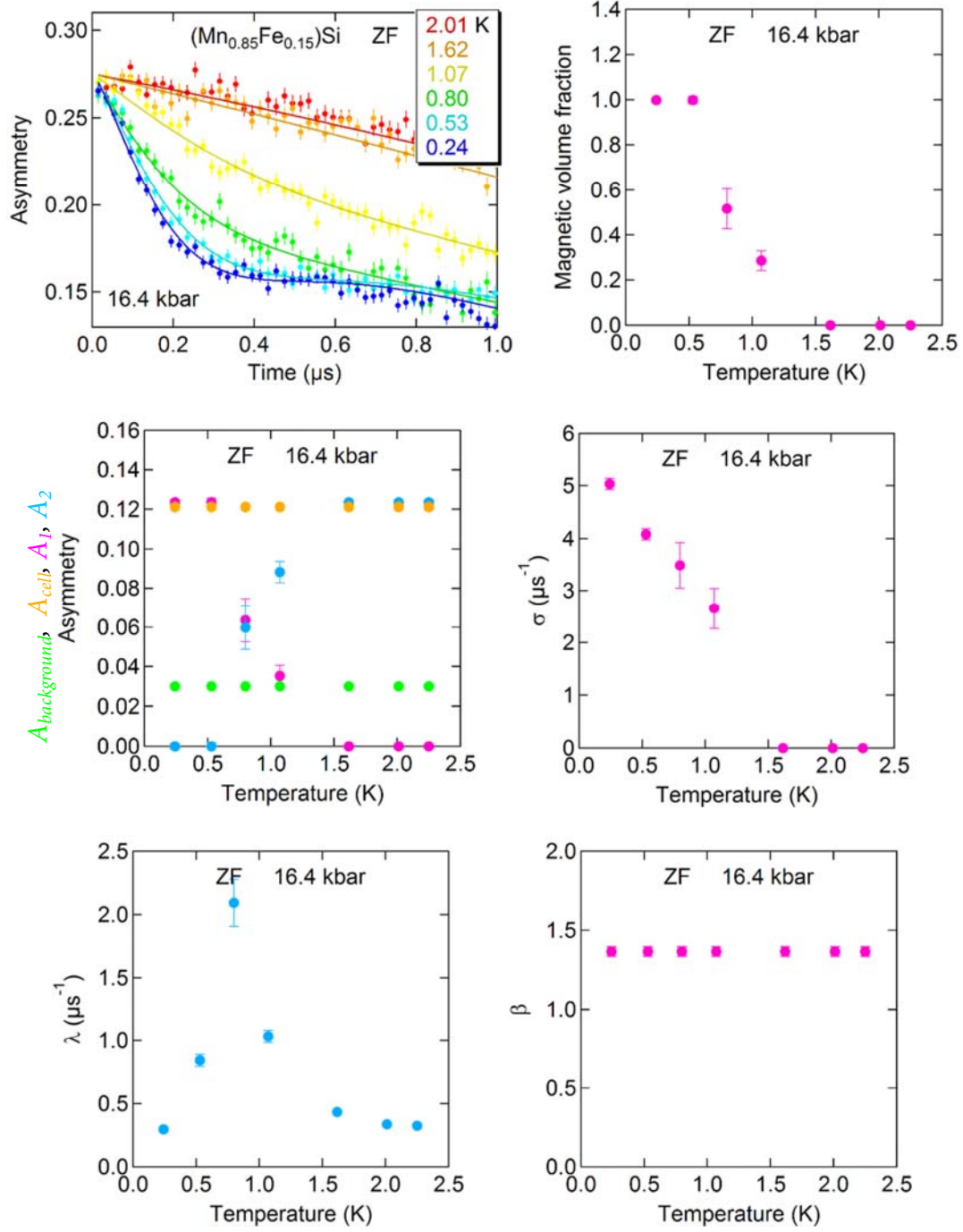
$$A_{\text{cell}} = 0.121$$

$$A_{\text{sample}} = A_{\text{total}} - A_{\text{cell}} - A_{\text{background}} = 0.124$$

$$A_{\text{sample}} : A_{\text{cell}} + A_{\text{background}} = 0.449 : 0.551$$

$$f_{\text{mag}} = \frac{A_1}{A_{\text{sample}}} = 8.09 \times A_1$$

(d) ZF 16.4 kbar



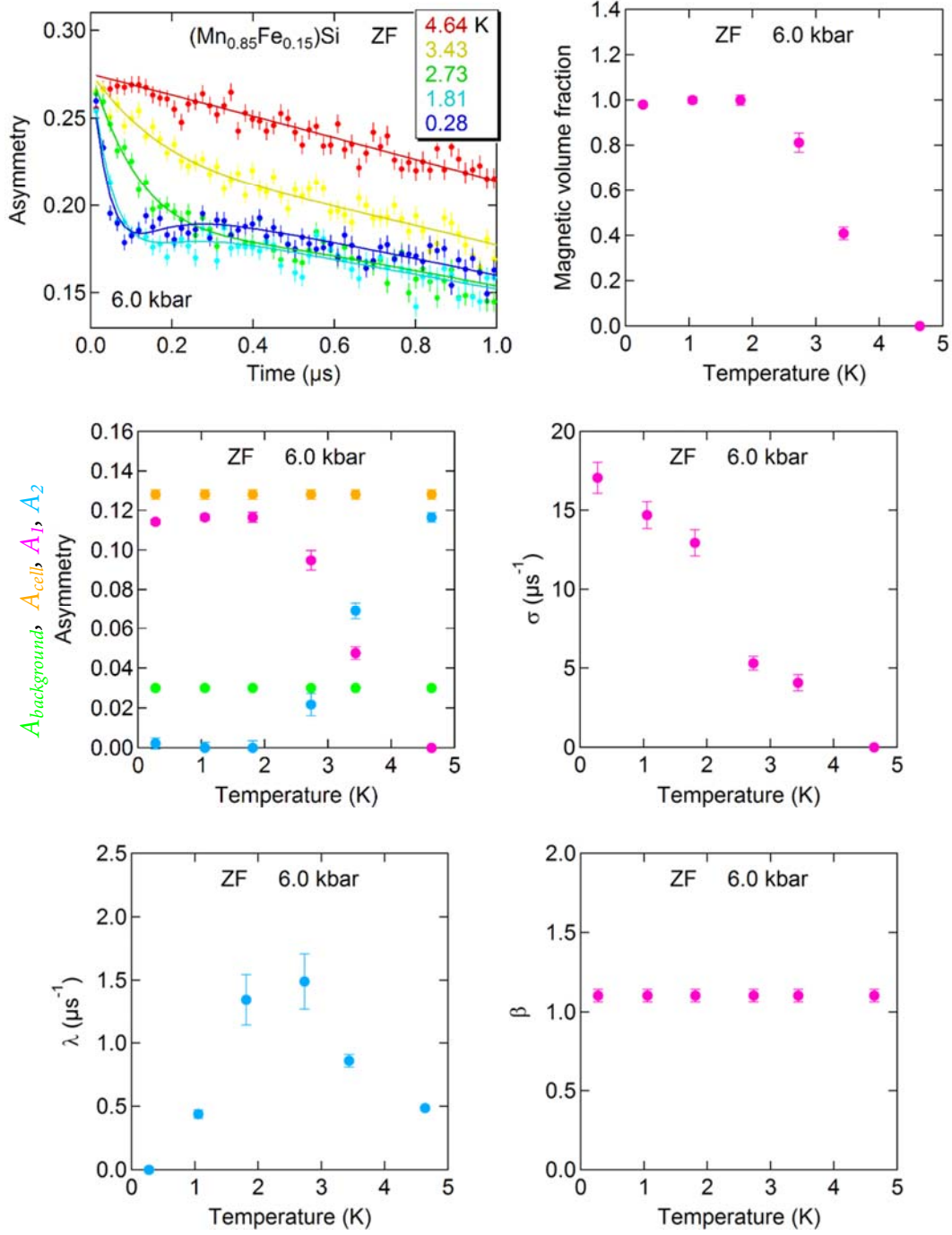
$$A_{\text{cell}} = 0.121$$

$$A_{\text{sample}} = A_{\text{total}} - A_{\text{cell}} - A_{\text{background}} = 0.124$$

$$A_{\text{sample}} : A_{\text{cell}} + A_{\text{background}} = 0.449 : 0.551$$

$$f_{\text{mag}} = \frac{A_1}{A_{\text{sample}}} = 8.09 \times A_1$$

(e) ZF 6.0 kbar



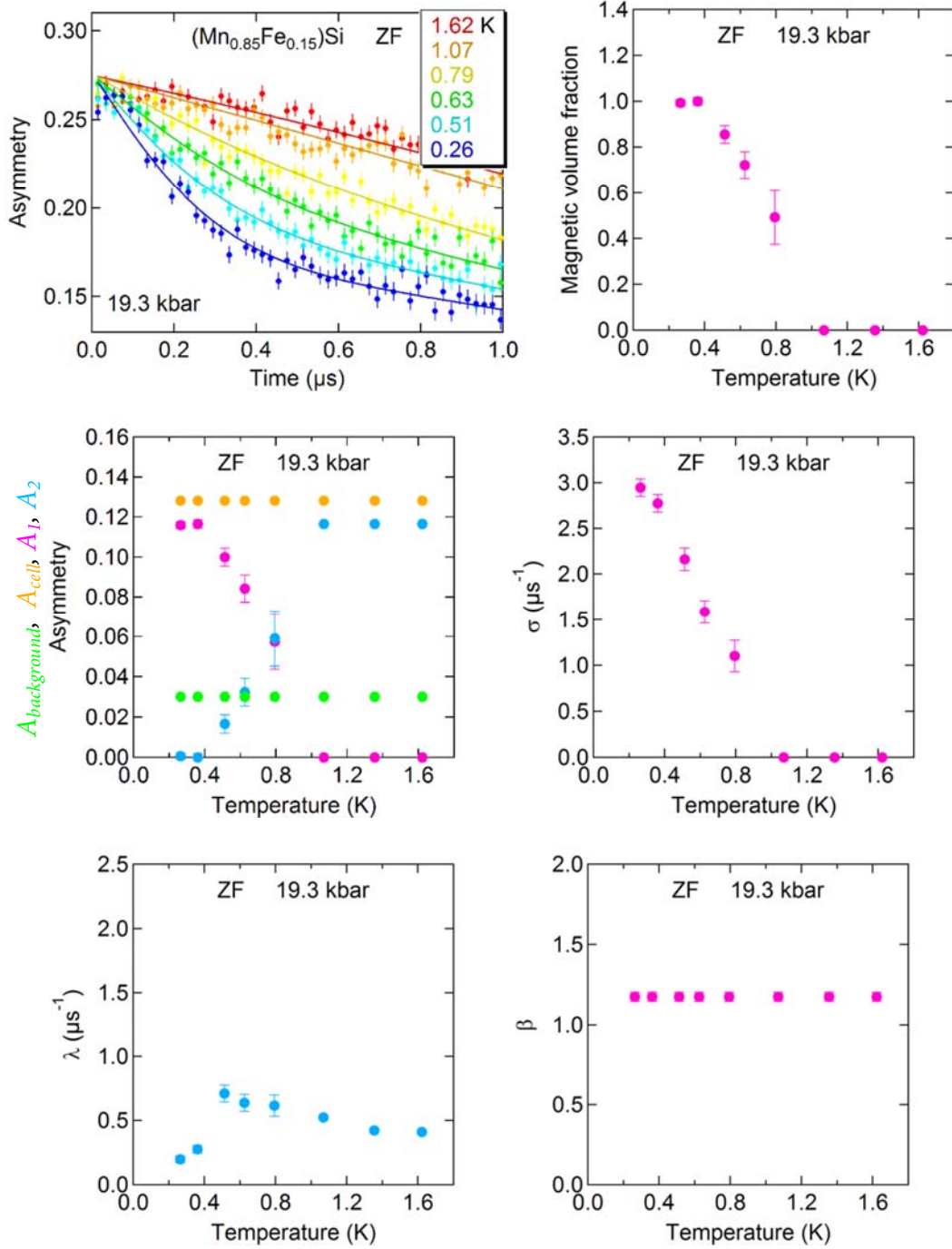
$$A_{\text{cell}} = 0.128$$

$$A_{\text{sample}} = A_{\text{total}} - A_{\text{cell}} - A_{\text{background}} = 0.117$$

$$A_{\text{sample}} : A_{\text{cell}} + A_{\text{background}} = 0.424 : 0.576$$

$$f_{\text{mag}} = \frac{A_1}{A_{\text{sample}}} = 8.57 \times A_1$$

(f) ZF 19.3 kbar



$$A_{\text{cell}} = 0.128$$

$$A_{\text{sample}} = A_{\text{total}} - A_{\text{cell}} - A_{\text{background}} = 0.117$$

$$A_{\text{sample}} : A_{\text{cell}} + A_{\text{background}} = 0.424 : 0.576$$

$$f_{\text{mag}} = \frac{A_1}{A_{\text{sample}}} = 8.57 \times A_1$$

(g) Empty Cell

$$A_1 \exp(-(\lambda t)^\beta) \left\{ \frac{1}{3} + \frac{2}{3} (1 - (\Delta t)^2) \exp\left(-\frac{(\Delta t)^2}{2}\right) \right\} + A_2$$

