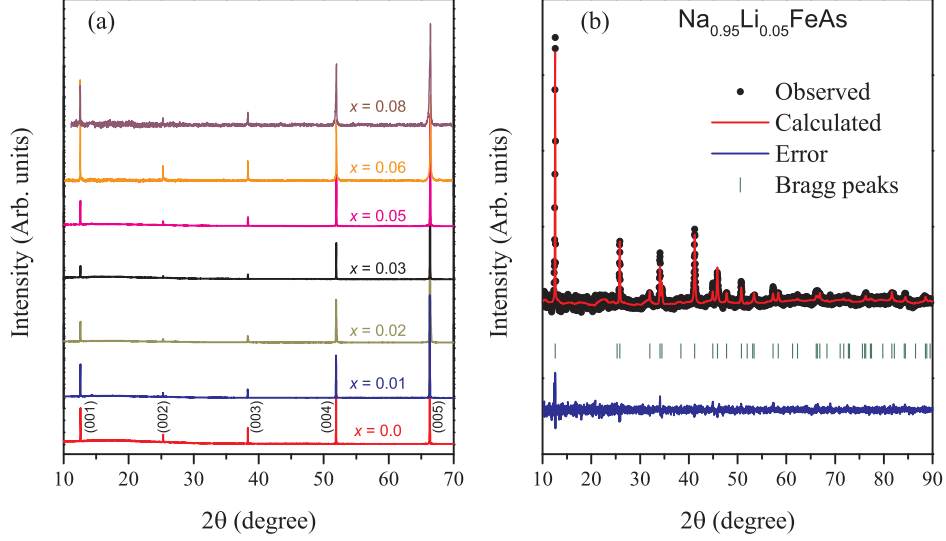
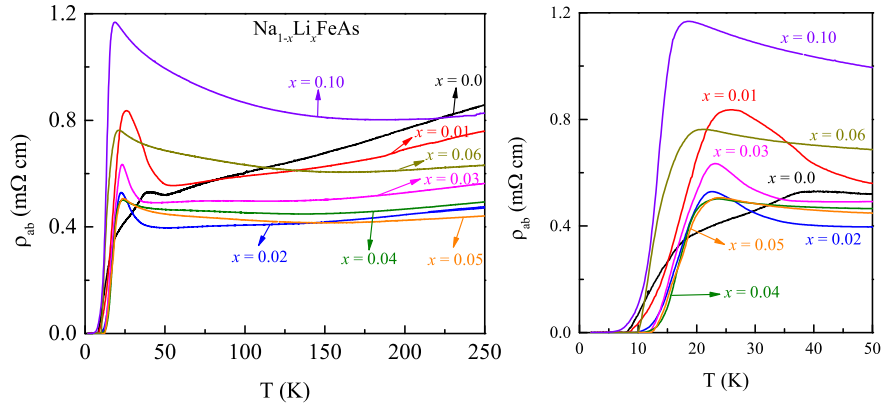
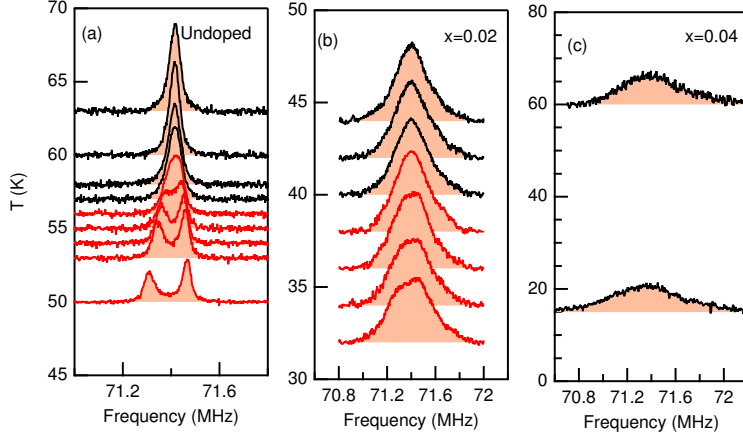




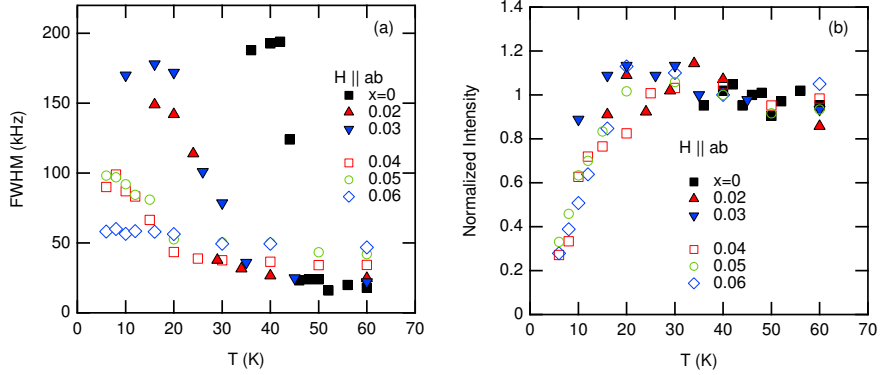
**Supplementary Figure 1.** X-ray diffraction data. (a) The single crystalline x-ray diffraction data of selected  $\text{Na}_{1-x}\text{Li}_x\text{FeAs}$  single crystals.  $(00l)$  reflection peaks can only be seen in the XRD patterns suggesting the absence of any other impurity phase. The diffraction patterns could be successfully refined by the tetragonal  $P4/nmm$  structure as in the parent  $\text{NaFeAs}$  (ref. 1) (b) Powder diffraction pattern of a ground  $\text{Na}_{0.95}\text{Li}_{0.05}\text{FeAs}$  single crystals.



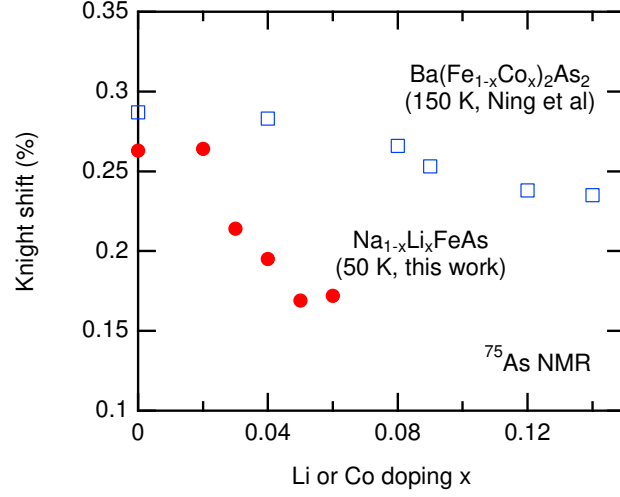
**Supplementary Figure 2.** Original resistivity data. Temperature dependence of the in-plane resistivity in  $\text{Na}_{1-x}\text{Li}_x\text{FeAs}$  single crystals for different Li concentration  $x$ .



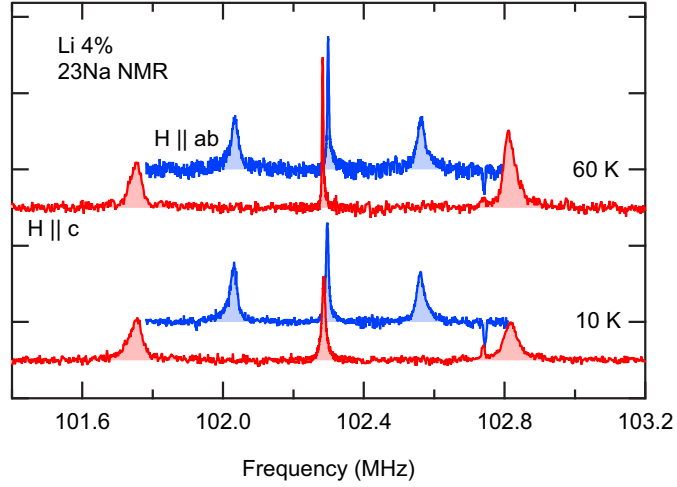
**Supplementary Figure 3.** Determination of the nematic transition temperature by NMR. Temperature dependence of the  $^{75}\text{As}$  satellite line for (a)  $x = 0$ , (b) 0.02, and (c) 0.04. The line splitting caused by nematic order was observed for  $x = 0$  and 0.02, which allows us to determine the nematic transition temperature. For  $x = 0.04$ , the splitting is not detected, probably due to the larger broadening of the line than the splitting.



**Supplementary Figure 4.** Detailed analysis of  $^{75}\text{As}$  spectra. (a) The full width at half maximum (FWHM) as a function of doping and temperature. For  $x \geq 0.04$ , the FWHM is weakly broadened down to low temperatures. Note that for  $x = 0.06$  there is no broadening of the line at all, while the sharp peak of  $(T_1T)^{-1}$  is clearly observed. For  $x = 0$ , the FWHM data were multiplied by 4 to compare directly with those of doped samples. (b) Normalized signal intensity as a function of doping and temperature. While the signal intensity does not change within experimental error for  $x < 0.03$ , it is notably suppressed below  $T_0$  for  $x \geq 0.04$ .



**Supplementary Figure 5.** Sharp reduction of the Knight shift near  $x = 0.03$ . Doping dependence of the Knight shift  $\mathcal{K}$  at 50 K is compared with data in  $\text{Ba(Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  at 150 K (taken from ref. 2).



**Supplementary Figure 6.** Comparison of  $^{23}\text{Na}$  spectra above and below  $T_0$  for  $x = 0.04$ . The spectrum does not reveal a notable difference in the ordered phase, unlike the  $^{75}\text{As}$  results, reflecting that the  $^{75}\text{As}$  effectively probes the Fe sites due to the much stronger hyperfine coupling.

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- <sup>1</sup> D. R. Parker, M. J. Pitcher, P. J. Baker, I. Franke, T. Lancaster, S. J. Blundell, and S. J. Clarke, “Structure, antiferromagnetism and superconductivity of the layered iron arsenide NaFeAs,” Chem. Commun. , 2189–2191 (2009).
- <sup>2</sup> F. L. Ning, K. Ahilan, T. Imai, A. S. Sefat, M. A. McGuire, B. C. Sales, D. Mandrus, P. Cheng, B. Shen, and H.-H Wen, “Contrasting Spin Dynamics between Underdoped and Overdoped Ba(Fe<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>As<sub>2</sub>,” Phys. Rev. Lett. **104**, 037001 (2010).