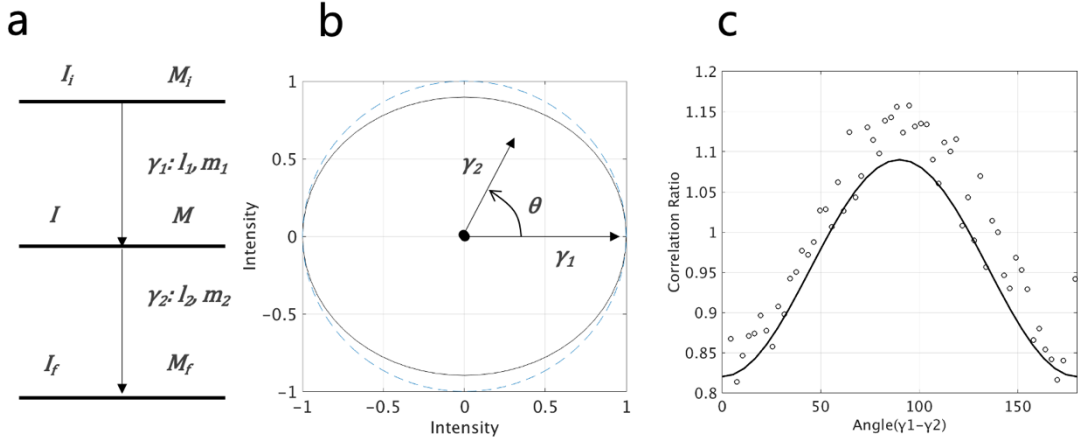
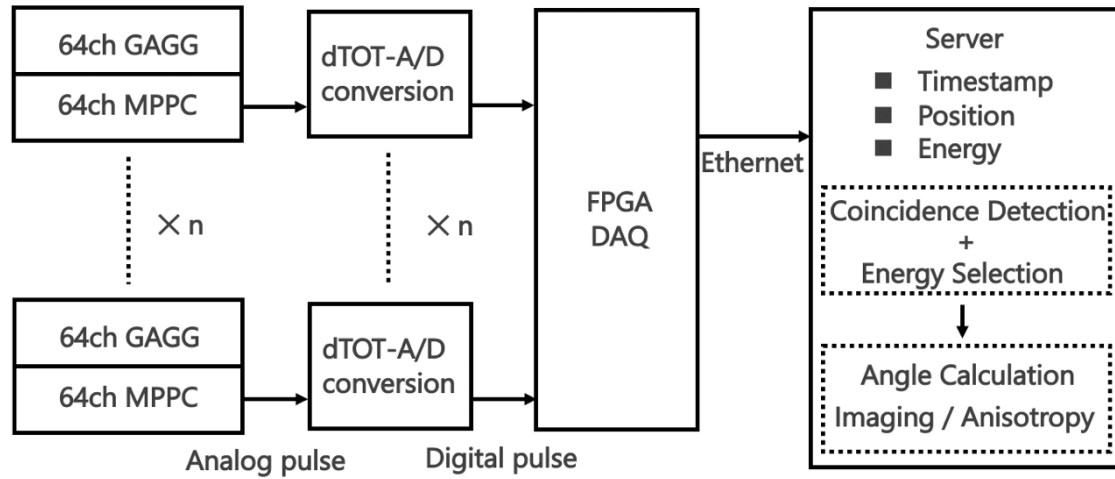


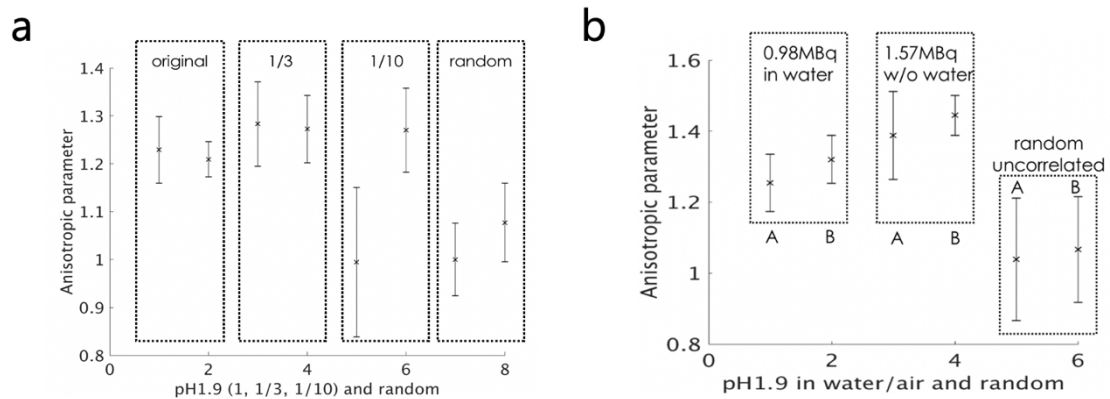
■ Supplementary figure/table legends



Supplementary Figure 1. Angular correlation and theoretical calculation. **a)** The generalized cascade decay, which has the initial state (I_i, M_i) , intermediate state (I, M) , and final state (I_f, M_f) , and emits two gamma-rays of $\gamma_1: l_1, m_1$ and $\gamma_2: l_2, m_2$, where I is nuclear spin, M is nuclear spin magnetic quantum number, l is angular momentum, and m is the magnetic quantum number of gamma-ray photons. **b)** The calculated anisotropy with Clebsch–Gordan coefficient for ^{111}In cascade decay, where θ is the angle from the first gamma-ray photon to the second one. **c)** Theoretical correlation ratio depending on the angle between two gamma-ray photons. Correlation ratio is calculated from the correlated intensity divided by isotropic emission. The circle dots show the Monte Carlo simulation results with the Geant4 tool kit.



Supplementary Figure 2. Schematic of signal processing and data processing from gamma-ray detectors to the reconstruction server. One module consists of a 64-channel GAGG scintillation crystal individually coupled to an MPPC photodetection array. Four or eight modules are used in the experiments and operated in parallel to acquire the gamma-ray distribution. The charge signals are converted to digital values with multi-channel dTOT analog-to-digital conversion and recorded by an FPGA-based DAQ system. All the data including timestamp, energy, and positions are transferred to the data server and used for image reconstruction or sensing by extracting the correlation ratio.



Supplementary Table 1. Measured pH values, radioactivity levels, and constituents of the mixed solutions.

pH value	Radioactivity (MBq)	Solution
1.181	1.12	$^{111}\text{InCl}_3 + \text{HCl} + \text{NaOH}$
1.889	0.843	Raw $^{111}\text{InCl}_3$
2.910	1.213	$^{111}\text{InCl}_3 + \text{pH9 phosphate buffer (PB)}$
5.032	1.037	$^{111}\text{InCl}_3 + \text{pH7PB}$
7.343	0.628	$^{111}\text{InCl}_3 + \text{NaOH} + \text{pH9PB} + \text{pH4PB}$
9.625	1.130	$^{111}\text{InCl}_3 + \text{NaOH} + \text{pH9PB} + \text{pH7PB}$
11.53	0.991	$^{111}\text{InCl}_3 + \text{NaOH} + \text{HCl}$