**S1 Text. Details of data reduction of quasielastic neutron scattering spectra**

To extract the quasielastic neutron scattering (QENS) spectra arising from the internal dynamics of Syn, the QENS spectra of the buffer were subtracted from those of the Syn solution sample, based on the incoherent scattering cross-section of the protein and the buffer.

Based on the amino acid sequence of Syn (UniProt identifier P37840), the chemical composition of Syn is C627H784D228N166O216S4, assuming that all the exchangeable hydrogen are replaced with deuterium, and without taking account of protonation and dissociation. The incoherent scattering cross-section inc of Syn can be calculated from this chemical composition and the incoherent scattering cross-section incatom of the atoms in the protein (incH = 79.9 barn or 10-24 cm2, incD = 2.04 barn, incC = 0.001 barn, incN = 0.49 barn, incO = 0.0 barn, and incS = 0.007 barn, taken from Table 2.2 in Ref. 37 in the main text), using the equation,

inc =  molatom*NA*iatom ,

atom

where molatom is the molar number of the atoms, and *NA* is the Avogadro's number. Summation was done over all atoms in the protein. For the monomeric state of Syn, where the concentration of Syn was 9.5 mg/ml, inc for Syn in unit volume (1 cm3) is calculated to be 0.0246 cm-1. Assuming that the partial specific volume of Syn is 0.73, the volume fraction of the solvent in the unit volume is calculated to be 1 0.73×9.5×10-3 ≈ 0.993. Assuming, then, that the density of D2O is 1.11 g/cm3 and using this volume fraction, the incoherent scattering cross-section of D2O in the solvent is calculated to be 0.135 cm-1. Note that although the solvent contains salts (10 mM Na2DPO4, 1.76 mM KD2PO4, 137 mM NaCl, and 2.7 mM KCl), the contribution of these salts to the incoherent scattering cross-section is negligible compared with that of D2O because the incoherent scattering cross-section of each salt is less than 10-3 cm-1 (Na2DPO4: 4.8×10-4 cm-1, KD2PO4: 1.6×10-4 cm-1, NaCl: 5.4×10-4 cm-1, and KCl: 8.6×10-6 cm-1). The fraction of the contribution of the solvent to the spectra of Syn in the monomeric state is thus 0.846 (= 0.135/(0.135+0.0246)). This value is used as a scaling factor to subtract the buffer spectra from the sample spectra.

A similar calculation for the fibril state of Syn, in which the concentration of Syn is 46 mg/ml, reveals that the incoherent scattering cross-section are 0.119 cm-1 and 0.132 cm-1 for Syn and D2O, respectively. The scaling factor for the subtraction of the buffer spectra is thus 0.526 (= 0.132/(0.132+0.119)).

The transmission T can be calculated from the equation, T = exp(-d), where  is the total scattering cross-section of the sample, and d is the thickness of the sample.  is the sum of the incoherent scattering cross-section (inc), the coherent scattering cross-section (coh), and the absorption cross-section (abs) of the sample. inc for Syn and D2O were calculated above. coh and abs for Syn and D2O can be calculated similarly using, instead of the values of incatom, the values of the coherent scattering cross-section cohatom of the atoms (cohH = 1.759 barn, cohD = 5.597 barn, cohC = 5.554 barn, cohN = 11.01 barn, cohO = 4.235 barn, and cohS = 1.019 barn) and those of the absorption cross-section absatom of the atoms (absH = 0.333 barn, absD = 0.0005 barn, absC = 1.90 barn, absN = 1.90 barn, absO = 0.0002 barn, and absS = 0.53 barn; the values of cohatom and absatom are taken from Table 2.2 in Ref. 19 in the main text).  (= incSyn + incD2O + cohSyn + cohD2O + absSyn + absD2O) are calculated to be 0.765 cm-1 and 0.678 cm-1 for the fibril-state sample and the monomeric-state sample, respectively. Using the sample thickness of 0.5 mm for the fibril-state sample and 1.0 mm for the monomeric-state sample, the transmission is calculated to be 0.962 and 0.934 for the fibril-state sample and the monomeric-state sample, respectively. The effect of multiple scattering should be negligible for samples with such high transmission (see S2 Text).