
1 **ARTICLE**

2 **Experimental validation of unique combination numbers for**
3 **third- and fourth-order neutron correlation factors of zero-power reactor noise**

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10 **Abstract**

11 Zero-power reactor noise is useful for subcriticality measurements. Based on the nuclear reactor
12 physics and the theory of neutron detection, this paper theoretically clarifies that the third- and
13 fourth-order neutron correlation factors \mathcal{Y}_3 and \mathcal{Y}_4 can be expressed as functions of the
14 second-order neutron correlation factor Y . In particular, if the neutron-counting gate width is
15 sufficiently large, the saturation values \mathcal{Y}_3/Y^2 and \mathcal{Y}_4/Y^3 are almost equal to the unique
16 combination numbers, ‘3’ and ‘15,’ for a source-driven subcritical system, where the
17 subcriticality is less than 10000 pcm. These unique combination numbers, ‘3’ and ‘15,’ for
18 \mathcal{Y}_3/Y^2 and \mathcal{Y}_4/Y^3 were validated using actual zero-power reactor noise measurements
19 carried out at the Kyoto University Criticality Assembly. In this study, the estimation of
20 statistical errors and correlations between different gate widths owing to the bunching method
21 was achieved by the moving block bootstrap method. For a sufficiently long measured reactor
22 noise in a steady and unperturbed state, a statistical test for the evaluation of the critical state
23 and the absolute measurement of subcriticality can be carried out by statistically quantifying
24 the difference between the measurement value of \mathcal{Y}_3/Y^2 and the unique combination number.

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3 *subcriticality; double factorial; bootstrap method; statistical error; covariance; KUCA;*

4 *measurement*

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1 **1. Introduction**

2 Research on subcriticality measurement techniques is important to experimentally ensure
3 criticality safety. Although various measurement techniques have been proposed and
4 implemented, each technique presents both advantages and disadvantages. For example, for an
5 unknown target system with a steady state, a dynamic technique such as the inverse kinetics
6 method [1,2,3] is not applicable. A static technique such as the neutron source multiplication
7 method [4,5,6] cannot be used to determine the absolute value of the effective neutron
8 multiplication factor k_{eff} for the target system without additional information; *e.g.*, the neutron
9 count rate for a reference state wherein k_{eff} is known, or the product of the detector efficiency
10 and effective source strength. If there is no information on the presence of an external neutron
11 source of which the strength cannot be neglected when compared with fission neutrons, the
12 evaluation of the critical state is not simple using the conventional static technique, which
13 focuses only on the average value of the neutron count rate in the target system.

14 In such a stationary unknown system, the measurement of the zero-power reactor noise (or
15 the fluctuation of the neutron count around the average value) is useful to estimate the
16 information related to the neutron multiplication [7]. The Feynman- α [8,9,10,11], Rossi- α
17 [12,13], and power spectral density [14,15] methods are used as reactor noise analysis
18 techniques, to measure the prompt neutron decay constant α that is expressed as $\alpha \approx$
19 $(\beta_{\text{eff}} - \rho)/\Lambda$ under a near-critical situation; where $-\rho \equiv (1 - k_{\text{eff}})/k_{\text{eff}}$, β_{eff} , and Λ are the
20 subcriticality, effective delayed neutron fraction, and neutron generation time, respectively. For
21 example, in the Feynman- α method, the time-series data of neutron counts are first measured.
22 The second-order neutron correlation factor Y is then evaluated from the variance-to-mean
23 ratio of the neutron counts, followed by the fitting procedure for the estimation of α . It should
24 be noted that the point kinetics parameters β_{eff} and Λ or the prompt neutron decay constant
25 at the critical state ($\alpha_{\text{crit}} = \beta_{\text{eff}}/\Lambda$) are required for the conversion from the measurement value
26 of α to the subcriticality $-\rho$.

1 Accordingly, in previous research, it was suggested that the third-order neutron correlation
2 factor \mathcal{Y}_3 contains useful information for the determination of the absolute value of the
3 subcriticality $-\rho$ [16,17,18]. However, few reports are available on the experimental studies
4 conducted on the higher-order neutron correlation factor [19,20]. One of the reasons for this is
5 that the precise measurement of the higher-order neutron correlation factor is difficult, because
6 the total measurement time of an experimental facility is limited. Moreover, an estimation
7 technique for the statistical error of the higher-order neutron correlation factor was not
8 sufficiently established in previous studies.

9 In recent studies, a technique using the bootstrap method [21,22] to estimate the statistical
10 error of the second-order neutron correlation factor Y was proposed [10,11]. The estimated
11 statistical error of Y was validated using the actual reactor noise measurement carried out at
12 the Kyoto University Critical Assembly (KUCA). This statistical error estimation technique
13 based on the bootstrap method can be also applied to the analog Monte Carlo simulation for the
14 reactor noise measurement [23].

15 The aim of this study was to extend the applicability of the bootstrap method, for the
16 practical estimation of the statistical errors of the third- and fourth-order neutron correlation
17 factors (\mathcal{Y}_3 and \mathcal{Y}_4), in addition to the covariance matrices between different neutron-counting
18 gate widths. Furthermore, by analyzing the reactor noise measurement carried out at the KUCA,
19 the fundamental physical properties of the third- and fourth-order neutron correlation factors in
20 the source-driven subcritical system were clarified. In particular, the primary aim was the
21 demonstration of the following relationships in the case wherein the neutron-counting gate
22 width T is sufficiently large under a relatively near-critical situation: $\mathcal{Y}_3/Y^2 \approx 3$ and
23 $\mathcal{Y}_4/Y^3 \approx 15$. As described in the later section, the specific numbers of ‘3’ and ‘15’ are referred
24 to as ‘unique combination numbers’ in this paper.

25 The remainder of the paper is structured as follows. In Section 2, a theory of the higher-
26 order neutron correlation factors is presented. In Section 3, an explanation of the bootstrap

1 method for the efficient estimation of statistical errors and correlations (covariance matrices)
2 of the higher-order neutron correlation factors is presented. Section 4 presents a simple
3 numerical simulation of the reactor noise for a non-multiplication system with a stationary
4 external neutron source, to better characterize the property of the reactor noise in the non-
5 multiplication system. In Section 5, the unique combination numbers of \mathcal{Y}_3/Y^2 and \mathcal{Y}_4/Y^3
6 are discussed with respect to an experimental analysis conducted on the actual zero-power
7 reactor noise data that were measured at the KUCA. Section 6 notes the focus of future work,
8 followed by the concluding remarks in Section 7.

9

10 **2. Theory**

11 **2.1. Higher-order neutron correlation factors**

12 Under the assumption that neutron counts $C(T)$ are detected within a counting gate width
13 T for a steady state of a source-driven subcritical system, the n th-order neutron correlation
14 factor $\mathcal{Y}_n(T)$ is defined as follows ($n \geq 2$):

$$\mathcal{Y}_n(T) \equiv \frac{1}{\langle C(T) \rangle} \frac{\partial^n}{\partial Z^n} \ln(G(Z, T)) \Big|_{Z=1}, \quad (1)$$

$$G(Z, T) \equiv \sum_{C=0}^{\infty} Z^C P(C, T), \quad (2)$$

15 where $P(C, T)$ is the probability that C neutrons are detected during the counting gate width
16 T owing to the stationary external neutron source; $G(Z, T)$ is the probability generating
17 function for $P(C, T)$; and the brackets $\langle \rangle$ indicate the expected value. For example, based on
18 Equation (1), the second-, third-, and fourth-order neutron correlation factors ($Y(T)$, $\mathcal{Y}_3(T)$,
19 and $\mathcal{Y}_4(T)$) can be evaluated as

$$Y(T) \equiv \mathcal{Y}_2(T) \equiv \frac{\kappa_2(T)}{\mu(T)} - 1, \quad (3)$$

$$\mathcal{Y}_3(T) \equiv \frac{\kappa_3(T)}{\mu(T)} - 3 \frac{\kappa_2(T)}{\mu(T)} + 2 = \left(\frac{\kappa_3(T)}{\mu(T)} - 1 \right) - 3 \left(\frac{\kappa_2(T)}{\mu(T)} - 1 \right), \quad (4)$$

$$\begin{aligned} \mathcal{Y}_4(T) &\equiv \frac{\kappa_4(T)}{\mu(T)} - 6 \frac{\kappa_3(T)}{\mu(T)} + 11 \frac{\kappa_2(T)}{\mu(T)} - 6 \\ &= \left(\frac{\kappa_4(T)}{\mu(T)} - 1 \right) - 6 \left(\frac{\kappa_3(T)}{\mu(T)} - 1 \right) + 11 \left(\frac{\kappa_2(T)}{\mu(T)} - 1 \right), \end{aligned} \quad (5)$$

1 where $\mu(T)$, $\kappa_2(T)$, $\kappa_3(T)$, and $\kappa_4(T)$ are the mean, and the second-, third-, and fourth-
2 order cumulants, which are respectively defined as

$$\mu(T) \equiv \langle C(T) \rangle, \quad (6)$$

$$\kappa_2(T) \equiv \langle (C(T) - \mu(T))^2 \rangle, \quad (7)$$

$$\kappa_3(T) \equiv \langle (C(T) - \mu(T))^3 \rangle, \quad (8)$$

$$\kappa_4(T) \equiv \langle (C(T) - \mu(T))^4 \rangle - 3(\kappa_2(T))^2. \quad (9)$$

3 It should be noted that $\kappa_2(T)$ is identical to the variance, and $\kappa_3(T)$ and $\kappa_4(T)$ correspond
4 to the skewness and kurtosis, respectively. If the probability distribution of $C(T)$ follows a
5 Poisson distribution, $\mu(T) = \kappa_2(T) = \kappa_3(T) = \kappa_4(T)$. Therefore, $Y(T)$, $\mathcal{Y}_3(T)$, and $\mathcal{Y}_4(T)$
6 represent measures of the relative deviation from the Poisson distribution.

7 In an actual reactor noise measurement, the total measurement time is limited; therefore,
8 the total number of samples for $C(T)$ is also finite. To reduce the bias for the limited number
9 of neutron count data [24], the unbiased estimators $k_2(T)$, $k_3(T)$, and $k_4(T)$ are used for the
10 experimental analysis of $\kappa_2(T)$, $\kappa_3(T)$, and $\kappa_4(T)$ [25]:

$$C_{\text{ave}}(T) = \frac{1}{N} \sum_{i=1}^N C_i(T), \quad (10)$$

$$k_2(T) = \frac{1}{N-1} \sum_{i=1}^N (C_i(T) - C_{\text{ave}}(T))^2, \quad (11)$$

$$k_3(T) = \frac{N}{(N-1)(N-2)} \sum_{i=1}^N (C_i(T) - C_{\text{ave}}(T))^3, \quad (12)$$

$$k_4(T) = \frac{N(N+1)}{(N-1)(N-2)(N-3)} \sum_{i=1}^N (C_i(T) - C_{\text{ave}}(T))^4 - \frac{3}{(N-2)(N-3)} \left(\sum_{i=1}^N (C_i(T) - C_{\text{ave}}(T))^2 \right)^2. \quad (13)$$

1 Consequently, the neutron correlation factors are experimentally estimated as:

$$Y(T) = \frac{k_2(T)}{C_{\text{ave}}(T)} - 1, \quad (14)$$

$$\mathcal{Y}_3(T) = \frac{k_3(T)}{C_{\text{ave}}(T)} - 3 \frac{k_2(T)}{C_{\text{ave}}(T)} + 2, \quad (15)$$

$$\mathcal{Y}_4(T) = \frac{k_4(T)}{C_{\text{ave}}(T)} - 6 \frac{k_3(T)}{C_{\text{ave}}(T)} + 11 \frac{k_2(T)}{C_{\text{ave}}(T)} - 6. \quad (16)$$

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3 **2.2. Saturation values for $Y(T)$, $\mathcal{Y}_3(T)$, and $\mathcal{Y}_4(T)$**

4 As reported in previous studies [11,26,27], analytical formulae for the neutron count rate
 5 R and saturation values of $Y(T)$, $\mathcal{Y}_3(T)$, and $\mathcal{Y}_4(T)$ in the limit of $T \rightarrow \infty$ can be derived
 6 using the first- to fourth-order detector importance functions, $I_1^\dagger(\vec{r}, E, \vec{\Omega})$ – $I_4^\dagger(\vec{r}, E, \vec{\Omega})$:

$$R \equiv \frac{\langle C(T) \rangle}{T} = \int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) q I_{1,s}^\dagger(\vec{r}) dV, \quad (17)$$

$$Y_\infty \equiv \lim_{T \rightarrow \infty} Y(T) = \frac{1}{R} \int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) \left(q I_{2,s}^\dagger(\vec{r}) + q(q-1) (I_{1,s}^\dagger(\vec{r}))^2 \right) dV, \quad (18)$$

$$\begin{aligned}
\mathcal{Y}_{3,\infty} &\equiv \lim_{T \rightarrow \infty} \mathcal{Y}_3(T) \\
&= \frac{1}{R} \int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) \left(\begin{array}{c} q \bar{I}_{3,s}^\dagger(\vec{r}) \\ +3q(q-1) \bar{I}_{1,s}^\dagger(\vec{r}) \bar{I}_{2,s}^\dagger(\vec{r}) \\ +q(q-1)(q-2) \left(\bar{I}_{1,s}^\dagger(\vec{r}) \right)^3 \end{array} \right) dV, \tag{19}
\end{aligned}$$

$$\begin{aligned}
\mathcal{Y}_{4,\infty} &\equiv \lim_{T \rightarrow \infty} \mathcal{Y}_4(T) \\
&= \frac{1}{R} \int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) \left(\begin{array}{c} q \bar{I}_{4,s}^\dagger(\vec{r}) \\ +4q(q-1) \bar{I}_{1,s}^\dagger(\vec{r}) \bar{I}_{3,s}^\dagger(\vec{r}) \\ +3q(q-1) \left(\bar{I}_{2,s}^\dagger(\vec{r}) \right)^2 \\ +6q(q-1)(q-2) \left(\bar{I}_{1,s}^\dagger(\vec{r}) \right)^2 \bar{I}_{2,s}^\dagger(\vec{r}) \\ +q(q-1)(q-2)(q-3) \left(\bar{I}_{1,s}^\dagger(\vec{r}) \right)^4 \end{array} \right) dV, \tag{20}
\end{aligned}$$

$$\bar{I}_{n,s}^\dagger(\vec{r}) \equiv \int_0^\infty dE' \int_{4\pi} d\Omega' \frac{\chi_s(\vec{r}, E')}{4\pi} I_n^\dagger(\vec{r}, E', \vec{\Omega}'), \tag{21}$$

1 where the subscript ‘ ∞ ’ indicates the saturation values in the limit of $T \rightarrow \infty$; $S(\vec{r})$ is the
2 spatial distribution of the source strength for the external neutron source; $\chi_s(\vec{r}, E)$ is the
3 energy spectrum of the external neutron source; $p_s(q, \vec{r})$ is the probability that q neutrons are
4 emitted per decay of the external source; and $\bar{I}_{n,s}^\dagger(\vec{r})$ is the weighted mean of the n th-order
5 detector importance function $I_n^\dagger(\vec{r}, E, \vec{\Omega})$, which satisfies the following adjoint neutron
6 transport equations [11,26,27]:

$$(\mathbf{A}^\dagger - \mathbf{F}^\dagger) I_1^\dagger(\vec{r}, E, \vec{\Omega}) = \Sigma_d(\vec{r}, E), \tag{22}$$

$$(\mathbf{A}^\dagger - \mathbf{F}^\dagger) I_2^\dagger(\vec{r}, E, \vec{\Omega}) = \Sigma_f(\vec{r}, E) \sum_{\nu=0}^{\infty} p_f(\nu, \vec{r}) \nu(\nu-1) \left(\bar{I}_{1,f}^\dagger(\vec{r}) \right)^2, \tag{23}$$

$$\begin{aligned}
&(\mathbf{A}^\dagger - \mathbf{F}^\dagger) I_3^\dagger(\vec{r}, E, \vec{\Omega}) \\
&= \Sigma_f(\vec{r}, E) \sum_{\nu=0}^{\infty} p_f(\nu, \vec{r}) \left(\begin{array}{c} 3\nu(\nu-1) \bar{I}_{1,f}^\dagger(\vec{r}) \bar{I}_{2,f}^\dagger(\vec{r}) \\ +\nu(\nu-1)(\nu-2) \left(\bar{I}_{1,f}^\dagger(\vec{r}) \right)^3 \end{array} \right), \tag{24}
\end{aligned}$$

$$\begin{aligned}
& (\mathbf{A}^\dagger - \mathbf{F}^\dagger) I_4^\dagger(\vec{r}, E, \vec{\Omega}) \\
&= \Sigma_f(\vec{r}, E) \sum_{\nu=0}^{\infty} p_f(\nu, \vec{r}) \left(\begin{array}{c} 4\nu(\nu-1) \bar{I}_{1,f}^\dagger(\vec{r}) \bar{I}_{3,f}^\dagger(\vec{r}) \\ + 3\nu(\nu-1) \left(\bar{I}_{2,f}^\dagger(\vec{r}) \right)^2 \\ + 6\nu(\nu-1)(\nu-2) \left(\bar{I}_{1,f}^\dagger(\vec{r}) \right)^2 \bar{I}_{2,f}^\dagger(\vec{r}) \\ + \nu(\nu-1)(\nu-2)(\nu-3) \left(\bar{I}_{1,f}^\dagger(\vec{r}) \right)^4 \end{array} \right), \quad (25)
\end{aligned}$$

$$\bar{I}_{n,f}^\dagger(\vec{r}) \equiv \int_0^\infty dE' \int_{4\pi} d\Omega' \frac{\chi_f(\vec{r}, E')}{4\pi} I_n^\dagger(\vec{r}, E', \vec{\Omega}'), \quad (26)$$

$$\mathbf{A}^\dagger \equiv -\vec{\Omega} \nabla + \Sigma_t(\vec{r}, E) - \int_0^\infty dE' \int_{4\pi} d\Omega' \Sigma_s(\vec{r}, E \rightarrow E', \vec{\Omega} \rightarrow \vec{\Omega}'), \quad (27)$$

$$\mathbf{F}^\dagger \equiv \nu \Sigma_f(\vec{r}, E) \int_0^\infty dE' \int_{4\pi} d\Omega' \frac{\chi_f(\vec{r}, E')}{4\pi}, \quad (28)$$

1 where the superscript ‘†’ indicates the adjoint; \mathbf{A}^\dagger and \mathbf{F}^\dagger are the adjoint net neutron loss,
2 and neutron production operators, respectively; $\Sigma_d(\vec{r}, E)$ is the macroscopic neutron detection
3 cross-section; $\chi_f(\vec{r}, E)$ is the energy spectrum of fission; $p_f(\nu, \vec{r})$ is the probability that ν
4 neutrons are emitted per fission; and the other notations maintain their conventional meanings
5 in the nuclear reactor physics.

6 Under the assumption that the fundamental mode approximation is applicable, $I_n^\dagger(\vec{r}, E, \vec{\Omega})$
7 is expressed as

$$I_n^\dagger(\vec{r}, E, \vec{\Omega}) \approx c_n \psi_0^\dagger(\vec{r}, E, \vec{\Omega}), \quad (29)$$

8 where c_n is the expansion coefficient, and $\psi_0^\dagger(\vec{r}, E, \vec{\Omega})$ is the adjoint k_{eff} -eigenfunction that
9 satisfies the following adjoint k_{eff} -eigenvalue equation:

$$\mathbf{A}^\dagger \psi_0^\dagger(\vec{r}, E, \vec{\Omega}) = \frac{1}{k_{\text{eff}}} \mathbf{F}^\dagger \psi_0^\dagger(\vec{r}, E, \vec{\Omega}). \quad (30)$$

10 The fundamental mode approximation of Equation (29) is more reasonable under a subcritical
11 condition, where the effective neutron multiplication factor k_{eff} is closer to unity. The
12 expansion coefficient c_n can be theoretically obtained by multiplying Equations (22)–(25) by

1 the forward k_{eff} -eigenfunction $\psi_0(\vec{r}, E, \vec{\Omega})$ and by using the orthogonality condition between
 2 the forward and adjoint eigenfunctions. Here, the forward eigenfunction $\psi_0(\vec{r}, E, \vec{\Omega})$ satisfies
 3 the following equation:

$$\mathbf{A}\psi_0(\vec{r}, E, \vec{\Omega}) = \frac{1}{k_{\text{eff}}}\mathbf{F}\psi_0(\vec{r}, E, \vec{\Omega}), \quad (31)$$

$$\mathbf{A} \equiv \vec{\Omega}\nabla + \Sigma_t(\vec{r}, E) - \int_0^\infty dE' \int_{4\pi} d\Omega' \Sigma_s(\vec{r}, E' \rightarrow E, \vec{\Omega}' \rightarrow \vec{\Omega}), \quad (32)$$

$$\mathbf{F} \equiv \frac{\chi_f(\vec{r}, E)}{4\pi} \int_0^\infty dE' \int_{4\pi} d\Omega' \nu \Sigma_f(\vec{r}, E'); \quad (33)$$

4 and the orthogonality condition of the eigenfunctions can be expressed as follows:

$$\int_V dV \int_0^\infty dE \int_{4\pi} d\Omega \left(\psi_0(\vec{r}, E, \vec{\Omega}) \mathbf{F}^\dagger \psi_m^\dagger(\vec{r}, E', \vec{\Omega}') \right) = \mathcal{F}_1 \delta_{0,m}, \quad (34)$$

5 where $\psi_m^\dagger(\vec{r}, E, \vec{\Omega})$ is the m th-order adjoint eigenfunction; $\delta_{0,m}$ is the Kronecker delta; and
 6 the parameter \mathcal{F}_n is conveniently introduced using the scalar flux ϕ_0 and the fission-source-
 7 averaged value for ψ_0^\dagger , as follows:

$$\mathcal{F}_n \equiv \int_V dV \int_0^\infty dE \Sigma_f(\vec{r}, E) \phi_0(\vec{r}, E) \sum_{\nu=0}^{\infty} \frac{\nu!}{(\nu-n)!} p_f(\nu, \vec{r}, E) \left(\bar{\psi}_{0,f}^\dagger(\vec{r}) \right)^n, \quad (35)$$

$$\phi_0(\vec{r}, E) \equiv \int_{4\pi} \psi_0(\vec{r}, E, \vec{\Omega}') d\Omega', \quad (36)$$

$$\bar{\psi}_{0,f}^\dagger(\vec{r}) \equiv \int_0^\infty dE' \int_{4\pi} d\Omega' \frac{\chi_f(\vec{r}, E')}{4\pi} \psi_0^\dagger(\vec{r}, E', \vec{\Omega}'). \quad (37)$$

8 Consequently, $I_n^\dagger(\vec{r}, E, \vec{\Omega})$ can be approximated as follows:

$$I_1^\dagger(\vec{r}, E, \vec{\Omega}) \approx \frac{\mathcal{D}}{-\rho\mathcal{F}_1} \psi_0^\dagger(\vec{r}, E, \vec{\Omega}), \quad (38)$$

$$I_2^\dagger(\vec{r}, E, \vec{\Omega}) \approx \left(\frac{\mathcal{D}}{-\rho\mathcal{F}_1} \right)^2 \frac{\mathcal{F}_2}{-\rho\mathcal{F}_1} \psi_0^\dagger(\vec{r}, E, \vec{\Omega}), \quad (39)$$

$$I_3^\dagger(\vec{r}, E, \vec{\Omega}) \approx \left(\frac{\mathcal{D}}{-\rho\mathcal{F}_1} \right)^3 \left(3 \left(\frac{\mathcal{F}_2}{-\rho\mathcal{F}_1} \right)^2 + \frac{\mathcal{F}_3}{-\rho\mathcal{F}_1} \right) \psi_0^\dagger(\vec{r}, E, \vec{\Omega}), \quad (40)$$

$$I_4^\dagger(\vec{r}, E, \vec{\Omega}) \approx \left(\frac{\mathcal{D}}{-\rho\mathcal{F}_1} \right)^4 \left(15 \left(\frac{\mathcal{F}_2}{-\rho\mathcal{F}_1} \right)^3 + 10 \frac{\mathcal{F}_2\mathcal{F}_3}{(-\rho\mathcal{F}_1)^2} + \frac{\mathcal{F}_4}{-\rho\mathcal{F}_1} \right) \psi_0^\dagger(\vec{r}, E, \vec{\Omega}), \quad (41)$$

$$\mathcal{D} \equiv \int_V dV \int_0^\infty dE \Sigma_d(\vec{r}, E) \phi_0(\vec{r}, E), \quad (42)$$

1 where the parameter \mathcal{D} is introduced for convenience and $\mathcal{D}/\mathcal{F}_1$ corresponds to the detection
2 efficiency.

3 By substituting Equations (38)–(41) into Equations (17)–(20), the saturation values based
4 on the fundamental mode approximation can be re-written as follows:

$$Y_\infty \approx \left(\frac{\mathcal{D}}{-\rho\mathcal{F}_1} \right) \left(\frac{\mathcal{F}_2}{-\rho\mathcal{F}_1} + \frac{\mathcal{S}_2}{\mathcal{S}_1} \right), \quad (43)$$

$$\mathcal{Y}_{3,\infty} \approx \left(\frac{\mathcal{D}}{-\rho\mathcal{F}_1} \right)^2 \left(\frac{\mathcal{F}_3}{-\rho\mathcal{F}_1} + \frac{\mathcal{S}_3}{\mathcal{S}_1} + 3 \frac{\mathcal{F}_2}{-\rho\mathcal{F}_1} \left(\frac{\mathcal{F}_2}{-\rho\mathcal{F}_1} + \frac{\mathcal{S}_2}{\mathcal{S}_1} \right) \right), \quad (44)$$

$$\mathcal{Y}_{4,\infty} \approx \left(\frac{\mathcal{D}}{-\rho\mathcal{F}_1} \right)^3 \left(\frac{\mathcal{F}_4}{-\rho\mathcal{F}_1} + \frac{\mathcal{S}_4}{\mathcal{S}_1} + 6 \frac{\mathcal{F}_2}{-\rho\mathcal{F}_1} \left(\frac{\mathcal{F}_3}{-\rho\mathcal{F}_1} + \frac{\mathcal{S}_3}{\mathcal{S}_1} \right) \right. \\ \left. + \left(4 \frac{\mathcal{F}_3}{-\rho\mathcal{F}_1} + 15 \left(\frac{\mathcal{F}_2}{-\rho\mathcal{F}_1} \right)^2 \right) \left(\frac{\mathcal{F}_2}{-\rho\mathcal{F}_1} + \frac{\mathcal{S}_2}{\mathcal{S}_1} \right) \right), \quad (45)$$

$$\mathcal{S}_n \equiv \int_V dV S(\vec{r}) \sum_{q=0}^{\infty} \frac{q!}{(q-n)!} p_s(q, \vec{r}) \left(\bar{\psi}_{0,s}^\dagger(\vec{r}) \right)^n, \quad (46)$$

$$\bar{\psi}_{0,s}^\dagger(\vec{r}) \equiv \int_0^\infty dE' \int_{4\pi} d\Omega' \frac{\chi_s(\vec{r}, E')}{4\pi} \psi_0^\dagger(\vec{r}, E', \vec{\Omega}'). \quad (47)$$

5 From Equations (43)–(45), if $-\rho < 0.1$ (dk/k) (or $0.9 < k_{\text{eff}} < 1$), the ratios of
6 $\mathcal{Y}_{3,\infty}/Y_\infty^2$ and $\mathcal{Y}_{4,\infty}/Y_\infty^3$ can be further approximated by linear functions with respect to $-\rho$:

$$\frac{\mathcal{Y}_{3,\infty}}{Y_\infty^2} \approx 3 + \frac{\mathcal{F}_1}{\mathcal{F}_2} \left(\frac{\mathcal{F}_3}{\mathcal{F}_2} - 3 \frac{\mathcal{S}_2}{\mathcal{S}_1} \right) (-\rho), \quad (48)$$

$$\frac{\mathcal{Y}_{4,\infty}}{Y_\infty^3} \approx 15 + 10 \frac{\mathcal{F}_1}{\mathcal{F}_2} \left(\frac{\mathcal{F}_3}{\mathcal{F}_2} - 3 \frac{\mathcal{S}_2}{\mathcal{S}_1} \right) (-\rho), \quad (49)$$

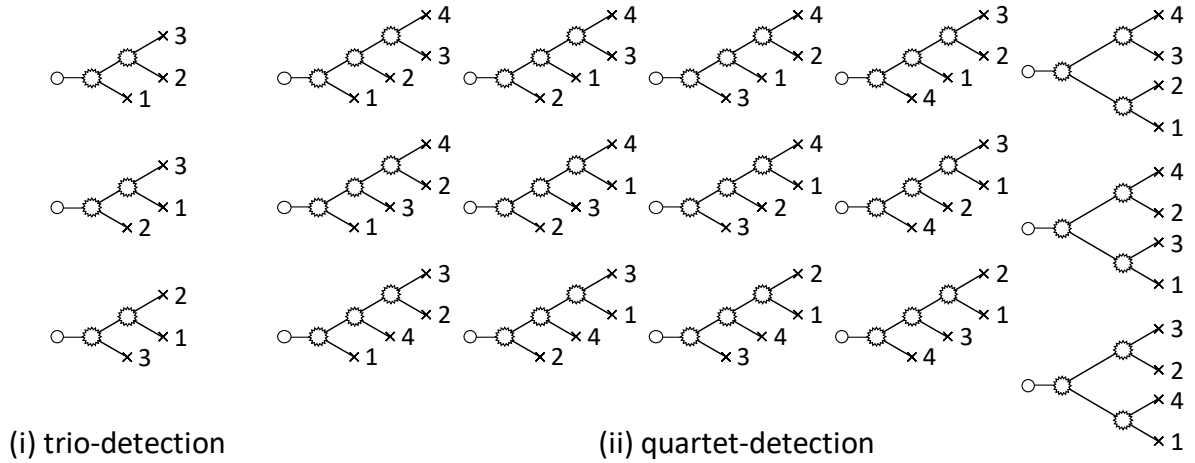
1 where the magnitude of $\left| \frac{\mathcal{F}_1}{\mathcal{F}_2} \left(\frac{\mathcal{F}_3}{\mathcal{F}_2} - 3 \frac{\mathcal{S}_2}{\mathcal{S}_1} \right) \right|$ is approximately 1 [18,28]. For a simple example,
 2 in the case of an infinite homogeneous system of ^{235}U with a Poisson source ($\langle q(q-1) \rangle = 0$),
 3 $\left| \frac{\mathcal{F}_1}{\mathcal{F}_2} \left(\frac{\mathcal{F}_3}{\mathcal{F}_2} - 3 \frac{\mathcal{S}_2}{\mathcal{S}_1} \right) \right| = \frac{\langle \nu \rangle \langle \nu(\nu-1)(\nu-2) \rangle}{\langle \nu(\nu-1) \rangle^2} \approx 0.8$. As k_{eff} approaches unity (or $-\rho \rightarrow +0$), $\mathcal{Y}_{3,\infty}/Y_\infty^2$
 4 and $\mathcal{Y}_{4,\infty}/Y_\infty^3$ converge to the constant values independent of \mathcal{F}_n and \mathcal{S}_n :

$$\lim_{-\rho \rightarrow +0} \frac{\mathcal{Y}_{3,\infty}}{Y_\infty^2} \approx 3, \quad (50)$$

$$\lim_{-\rho \rightarrow +0} \frac{\mathcal{Y}_{4,\infty}}{Y_\infty^3} \approx 15. \quad (51)$$

5 As shown in Figure 1, the unique combination numbers, ‘3’ and ‘15,’ correspond to the total
 6 number of combinations for the trio- and quartet-detections that have two and three two-forked
 7 branches, respectively.

8



9

10 **Figure 1** Combinations for trio- and quartet-detections

11

12 On the contrary, in the limit of $k_{\text{eff}} \rightarrow +0$ or $-\rho \rightarrow +\infty$, the higher-order importance
 13 functions $I_2^\dagger(\vec{r}, E, \vec{\Omega}) - I_4^\dagger(\vec{r}, E, \vec{\Omega})$ are zero, because $\Sigma_f(\vec{r}, E) = 0$ in Equations (23)–(25).
 14 Consequently, the ratios of $\mathcal{Y}_{3,\infty}/Y_\infty^2$ and $\mathcal{Y}_{4,\infty}/Y_\infty^3$ converge to particular values that are

1 dependent on the multiplicity of the external neutron source:

$$\lim_{-\rho \rightarrow +\infty} \frac{y_{3,\infty}}{Y_\infty^2} = \frac{\left(\left(\int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) q \bar{I}_{1,s}^+(\vec{r}) dV \right) \times \left(\int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) q(q-1)(q-2) \left(\bar{I}_{1,s}^+(\vec{r}) \right)^3 dV \right) \right)}{\left(\int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) q(q-1) \left(\bar{I}_{1,s}^+(\vec{r}) \right)^2 dV \right)^2}, \quad (52)$$

$$\lim_{-\rho \rightarrow +\infty} \frac{y_{4,\infty}}{Y_\infty^3} = \frac{\left(\left(\int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) q \bar{I}_{1,s}^+(\vec{r}) dV \right)^2 \times \left(\int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) q(q-1)(q-2)(q-3) \left(\bar{I}_{1,s}^+(\vec{r}) \right)^4 dV \right) \right)}{\left(\int_V S(\vec{r}) \sum_{q=0}^{\infty} p_s(q, \vec{r}) q(q-1) \left(\bar{I}_{1,s}^+(\vec{r}) \right)^2 dV \right)^3}. \quad (53)$$

2 If the external source $S(\vec{r})$ is a point-wise source of the Dirac delta function, Equations (52)–

3 (53) are further simplified as follows:

$$\lim_{-\rho \rightarrow +\infty} \frac{y_{3,\infty}}{Y_\infty^2} \approx \frac{\langle q \rangle \langle q(q-1)(q-2) \rangle}{\langle q(q-1) \rangle^2}, \quad (54)$$

$$\lim_{-\rho \rightarrow +\infty} \frac{y_{4,\infty}}{Y_\infty^3} \approx \frac{\langle q \rangle^2 \langle q(q-1)(q-2)(q-3) \rangle}{\langle q(q-1) \rangle^3}. \quad (55)$$

4 For example, in the case of a spontaneous fission nuclide such as ^{252}Cf , $\frac{\langle q \rangle \langle q(q-1)(q-2) \rangle}{\langle q(q-1) \rangle^2} \approx 0.8$

5 and $\frac{\langle q \rangle^2 \langle q(q-1)(q-2)(q-3) \rangle}{\langle q(q-1) \rangle^3} \approx 0.6$ [28], thus the ratios of $y_{3,\infty}/Y_\infty^2$ and $y_{4,\infty}/Y_\infty^3$ under the

6 condition of $k_{\text{eff}} = 0$ are significantly different from the unique combination numbers, ‘3’ and

7 ‘15,’ near the critical state.

8

9 **3. Bootstrap method for statistical error estimation of y_n**

10 In the previous study, the statistical error estimation for the Feynman- α method using the

11 bootstrap method was proposed for the practical estimation of the statistical errors of both Y

12 and the prompt neutron decay constant α [10]. Subsequently, the error estimation technique

13 was improved to effectively calculate the covariance matrix of $Y(T)$ between different gate

14 widths T [11]. This improvement was achieved using the recursive bunching method. In this

15 study, the moving block bootstrap method [22] was used to estimate the statistical errors of the

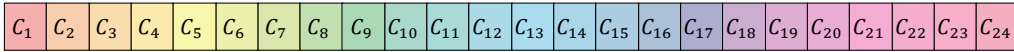
1 higher-order neutron correlation factors $\mathcal{Y}_n(T)$ and their covariance matrices. Details of the
 2 procedure are presented below:

- 3 1. The original time-series data of the neutron counts $\vec{C}(T_0) = [C_1, C_2, \dots, C_{N_0}]$ are provided
 4 by a single measurement of the reactor noise, where the basic counting gate width is T_0 , and
 5 the total number of count data is N_0 .
- 6 2. An upper limit value of the bunching is set as M , where $1 < M < N_0$.
- 7 3. An empty vector $\vec{C}^*(T_0) = []$ is prepared ($i = 1$).
- 8 4. The ‘resampling position ξ_i ’ is determined using a uniform random integer number, $1 \leq$
 9 $\xi_i \leq (N_0 - M + 1)$. Successive time-series data $\vec{C}_{\xi_i} = [C_{\xi_i}, C_{\xi_i+1}, \dots, C_{\xi_i+M-1}]$ are then
 10 extracted from the original time series data and added to the end of the vector $\vec{C}^*(T_0)$. This
 11 extraction of successive data is necessary for the estimate of the covariance matrices of the
 12 higher-order neutron correlation factors and the ratios.
- 13 5. As shown in Figure 2, a ‘bootstrap sample of the time-series data $\vec{C}^*(T_0)$ ’ is newly generated
 14 by repeating Step 4 L times:

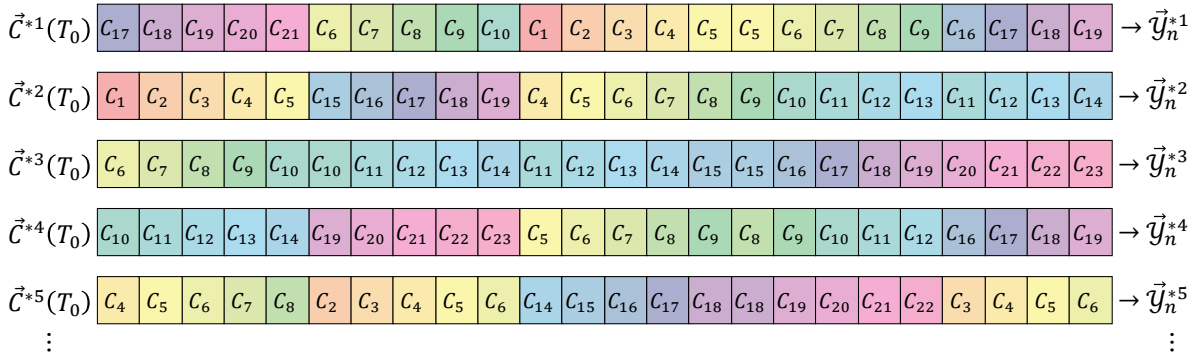
$$\vec{C}^*(T_0) = [\vec{C}_{\xi_1}, \vec{C}_{\xi_2}, \dots, \vec{C}_{\xi_L}], \quad (56)$$

15 where $L = [N_0/M]$. It should be noted that extra data in \vec{C}_{ξ_L} is removed if necessary, so
 16 that the total number of count data in $\vec{C}^*(T_0)$ is N_0 .

(a) original time-series data $C(T_0)$



(b) bootstrap method



17

18 **Figure 2** Example of the moving block bootstrap method ($N_0 = 24, M = 5$)

1

2 6. Using the recursive bunching method with Equations (10)–(16) for the bootstrap sample

3 $\vec{C}^*(T_0)$ in Step 5, the variations in the ‘bootstrap replicates $\mathcal{Y}_n^*(kT_0)$, $\frac{y_3^*(kT_0)}{(Y^*(kT_0))^2}$, and

4 $\frac{y_4^*(kT_0)}{(Y^*(kT_0))^3}$, are evaluated for the bunching gate width kT_0 , where k is the bunching

5 number ($1 \leq k \leq M$). As shown in Figure 3, the bunching method is recursively applied

6 to previously-bunched data, *i.e.*, $\vec{C}^*(2kT_0)$ is effectively produced by combining a pair

7 of successive elements in $\vec{C}^*(kT_0)$. Here, the bunching number k is given by $k = p \times$

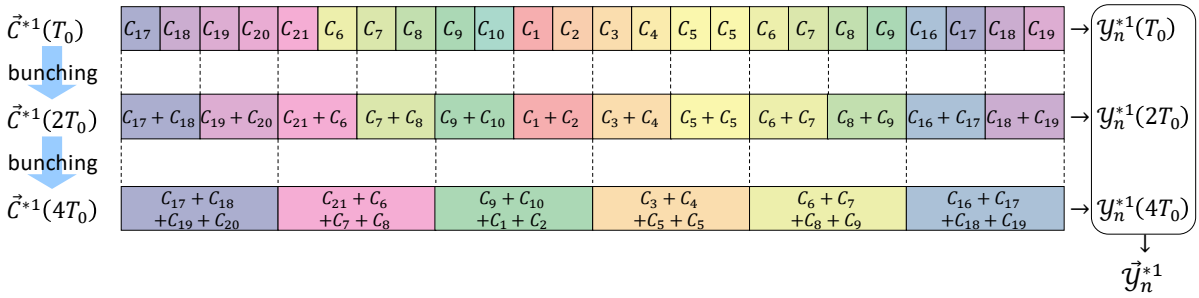
8 2^j ($j = 0, 1, \dots$). For example, the initial bunching number p is empirically determined

9 as $p = 2, 3, 5, 7, 9, 11, 13, 15, 17, 19, 21, 23, 25, 27, 29, 31$, and 33. The maximum value of j

10 is limited to satisfy $p \times 2^j \leq M$. Consequently, row vectors $\vec{y}_n^* =$

11 $[\mathcal{Y}_n^*(T_0), \mathcal{Y}_n^*(2T_0), \dots, \mathcal{Y}_n^*(MT_0)]$, $\vec{r}_3^* = \left[\frac{y_3^*(T_0)}{(Y^*(T_0))^2}, \frac{y_3^*(2T_0)}{(Y^*(2T_0))^2}, \dots, \frac{y_3^*(MT_0)}{(Y^*(MT_0))^2} \right]$, and $\vec{r}_4^* =$

12 $\left[\frac{y_4^*(T_0)}{(Y^*(T_0))^3}, \frac{y_4^*(2T_0)}{(Y^*(2T_0))^3}, \dots, \frac{y_4^*(MT_0)}{(Y^*(MT_0))^3} \right]$ are obtained.



13

14 **Figure 3** Example of the recursive bunching method ($p = 2$)

15

16 7. To estimate the confidence intervals and covariance matrices of \vec{y}_n^* , \vec{r}_3^* , and \vec{r}_4^* , Steps 3–

17 6 are repeated B times. Consequently, a set of bootstrap replicates \vec{y}_n^{*b} , \vec{r}_3^{*b} , and \vec{r}_4^{*b} ,

18 are obtained for $b = 1, 2, \dots, B$, where B is the total number of bootstrap replicates.

19 8. As a result of Step 7, frequency distributions of $\mathcal{Y}_n^*(kT_0)$, $\frac{y_3^*(kT_0)}{(Y^*(kT_0))^2}$, and $\frac{y_4^*(kT_0)}{(Y^*(kT_0))^3}$ are

20 obtained. On the basis of these ‘bootstrap frequency distributions,’ the percentile

1 confidence intervals (or 2.5 and 97.5 percentile points) can be simply estimated to evaluate
 2 the range of statistical errors of $\mathcal{Y}_n^*(kT_0)$, $\frac{\mathcal{Y}_3^*(kT_0)}{(Y^*(kT_0))^2}$, and $\frac{\mathcal{Y}_4^*(kT_0)}{(Y^*(kT_0))^3}$. For example, the B
 3 bootstrap replicates $\mathcal{Y}_n^{*b}(kT_0)$ are sorted in ascending order. The lower and upper limits
 4 of the 95% bootstrap confidence interval are simply estimated from the $(0.025 \times B)$ th
 5 and $(0.975 \times B)$ th smallest values of sorted $\mathcal{Y}_n^{*b}(kT_0)$, respectively.

6 9. Using the row vectors $\vec{\mathcal{Y}}_n^{*b}$, \vec{r}_3^{*b} , and \vec{r}_4^{*b} , these bootstrap covariance matrices can be
 7 estimated, if necessary. For example, the covariance matrix $\Sigma_{\mathcal{Y}_n^*}$ is calculated as follows:

$$\Sigma_{\mathcal{Y}_n^*} = \frac{1}{B-1} \sum_{b=1}^B (\vec{\mathcal{Y}}_n^{*b} - \vec{\mathcal{Y}}_{\text{ave}}^*)^T (\vec{\mathcal{Y}}_n^{*b} - \vec{\mathcal{Y}}_{\text{ave}}^*), \quad (57)$$

$$\vec{\mathcal{Y}}_{\text{ave}}^* = \frac{1}{B} \sum_{b=1}^B \vec{\mathcal{Y}}_n^{*b}, \quad (58)$$

8 where the superscript T indicates the transpose. In the same manner as presented in
 9 Equation (57), the covariance matrices $\Sigma_{r_3^*}$ and $\Sigma_{r_4^*}$ can be also estimated using \vec{r}_3^{*b}
 10 and \vec{r}_4^{*b} instead of $\vec{\mathcal{Y}}_n^{*b}$, respectively.

11

12 4. Zero-power reactor noise simulation for non-multiplication system

13 4.1. Calculation conditions of numerical simulation

14 To understand the reactor noise in the non-multiplication system, a very simple Monte
 15 Carlo simulation was conducted in the same manner as in the previous studies [24,27]. If
 16 possible, the validation should be carried out using an actual reactor noise measurement.
 17 However, it is not easy to measure the statistically significant data for an actual non-
 18 multiplication system with an external neutron source such as a ^{252}Cf spontaneous source, given
 19 that the magnitude of neutron correlation factors \mathcal{Y}_n are significantly small owing to the
 20 absence of fission chain reactions [29]. Thus, in future work, the validation will be conducted
 21 by carrying out actual measurements using a high-efficiency neutron detection system, which
 22 is beyond the scope of this study.

1 In the Monte Carlo simulation, an infinite homogenous system with a stationary external
2 neutron source was assumed. In this study, the external neutron source was a spontaneous
3 fission source of ^{252}Cf and the probability distribution of $p_s(q)$ was quoted from Reference
4 [28]. Therefore, the factorial moments of q were calculated as follows: $\langle q \rangle = 3.77$,
5 $\langle q(q-1) \rangle = 12.05$, $\langle q(q-1)(q-2) \rangle = 32.03$, and $\langle q(q-1)(q-2)(q-3) \rangle = 69.52$.
6 The source strength S was set as $S = 100$ (neutrons/s). For simplicity, the neutron energy
7 was one group, and all the absorbed neutrons were assumed to be detected to obtain the time-
8 series data. The product of the neutron velocity and macroscopic absorption cross-section ($v\Sigma_a$)
9 was set as $v\Sigma_a = 10000$ (1/s), which also corresponds to the prompt neutron decay constant
10 α in this simple problem. The virtual measurement time of the reactor noise (N_0T_0) was 10000
11 (s).

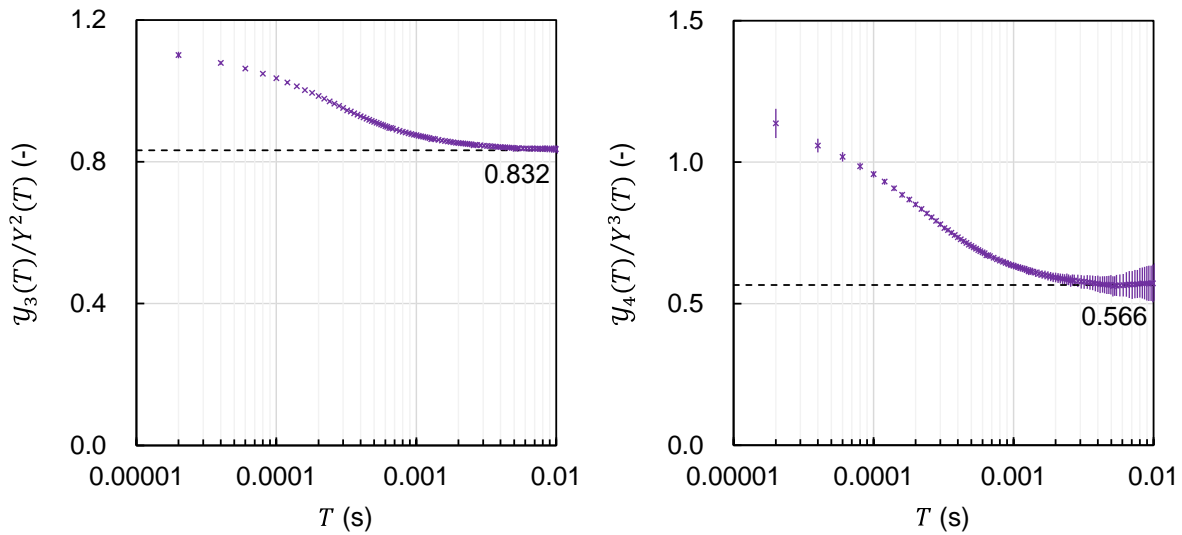
12 Using the recursive bunching method for the simulated time-series data, the saturation
13 values of $\frac{y_3(kT_0)}{(Y(kT_0))^2}$ and $\frac{y_4(kT_0)}{(Y(kT_0))^3}$ were numerically investigated using the methodology
14 described in Section 3, where $T_0 = 2 \times 10^{-5}$ (s), $M = 512$, and the total number of count
15 data N_0 was 5×10^8 . To estimate the statistical errors of $\frac{y_3(kT_0)}{(Y(kT_0))^2}$ and $\frac{y_4(kT_0)}{(Y(kT_0))^3}$, the 95%
16 bootstrap confidence intervals were also calculated using the moving block bootstrap method
17 with $B = 1000$.

18

19 4.2. Numerical results

20 Figure 4 presents numerical results of the ratios of $\frac{y_3(T)}{(Y(T))^2}$ and $\frac{y_4(T)}{(Y(T))^3}$, where the error
21 bars indicate the 95% bootstrap confidence intervals. As T becomes sufficiently large, it was
22 confirmed that these values converge to the saturation values. As can be expected with respect
23 to Equations (54) and (55), the saturation values were approximately $\frac{\langle q \rangle \langle q(q-1)(q-2) \rangle}{\langle q(q-1) \rangle^2} = 0.832$
24 and $\frac{\langle q \rangle^2 \langle q(q-1)(q-2)(q-3) \rangle}{\langle q(q-1) \rangle^3} = 0.566$, although the statistical errors tended to increase as T
25 increased. The saturation values of $\frac{y_3(T)}{(Y(T))^2}$ and $\frac{y_4(T)}{(Y(T))^3}$ in the non-multiplication system were
26 significantly different from the unique combination numbers, ‘3’ and ‘15,’ in the source-driven

1 subcritical system. This implies that the detection of the neutron multiplication induced by
 2 fissile materials can be statistically evaluated from the differences between $y_{3,\infty}/Y_{\infty}^2$ and '3'
 3 and between $y_{4,\infty}/Y_{\infty}^3$ and '15.'



4

5 **Figure 4** Numerical results of saturation values for $y_3(T)/(Y(T))^2$ and $y_4(T)/(Y(T))^3$
 6 in the non-multiplication system with ^{252}Cf source

7

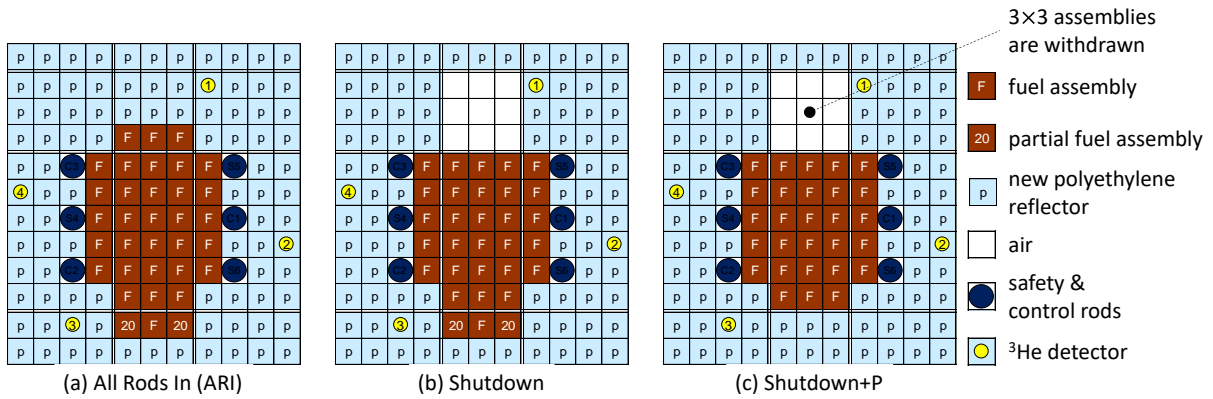
8 **5. Zero-power reactor noise measurements for actual subcritical system**

9 **5.1. Experimental conditions**

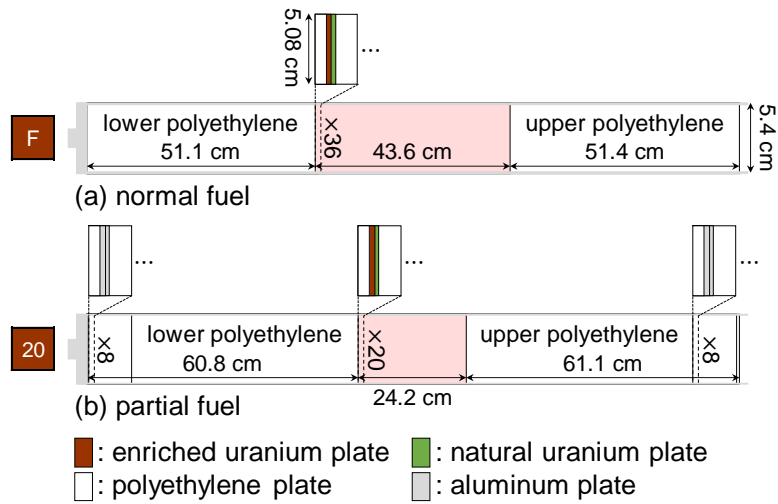
10 In the previous studies [10,11,30], several series of reactor noise experiments were
 11 conducted in the A-core (A3/8”p36EU-NU) at the KUCA. The experimental conditions are
 12 briefly explained below.

13 The experimental cores and the loaded fuel assemblies are presented in Figure 5 and Figure
 14 6, respectively. The core-average ^{235}U enrichment was 5.4 wt%. In the experimental analysis,
 15 the following three cases were analyzed: (a) All control Rods In (ARI), (b) Shutdown, and (c)
 16 Shutdown with the replacement of fuel assemblies using Polyethylene reflectors (Shutdown+P).
 17 In Cases (b) and (c), 3×3 fuel and reflector assemblies were fully withdrawn, because the reactor
 18 is shutdown state. In Case (c), the three fuel assemblies were replaced by the polyethylene

1 reflector assemblies to obtain the deeper subcriticality. For Cases (a)–(c), the numerical results
 2 of k_{eff} , β_{eff} , and Λ using MCNP6.2 [31] with JENDL-4.0 [32] are presented in Table 1.



3
 4 **Figure 5** Top view of experimental cores (A3/8”p36EU-NU)



6
 7 **Figure 6** Fuel assemblies loaded in experimental core

8
 9 **Table 1** Numerical results of neutronics parameters for experimental cores

Case	k_{eff} (-)	β_{eff} (pcm)	Λ (μs)
(a) ARI	0.98120±0.00003†	776±6	39.65±0.03
(b) Shutdown	0.95118±0.00003	789±6	41.73±0.04
(c) Shutdown+P	0.93589±0.00003	794±6	42.15±0.04

10 † : 1 σ statistical error

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In this experiment, four ^3He detectors (#1–4) were placed at the axial center positions of the ex-core reflector assemblies. Using these detectors with a list-mode data acquisition system, the time-series data of neutron counts were successively measured. In Cases (a)–(c), the reactor noise was measured without any external neutron source such as an Am-Be or Cf source. In other words, the measurement was carried out using only the inherent neutron source, which mainly consists of the spontaneous fission of ^{238}U and (α,n) reactions of ^{27}Al owing to the α -decay of uranium isotopes [30]. To increase the neutron count rate, all the time-series data using detectors #1–4 were summed for the reactor noise analysis. Thereby, the neutron count rates $R = C_{\text{ave}}(T)/T$ for Cases (a)–(c) were 74.12 ± 0.14 , 29.98 ± 0.04 , and 21.13 ± 0.03 (count/s), respectively.

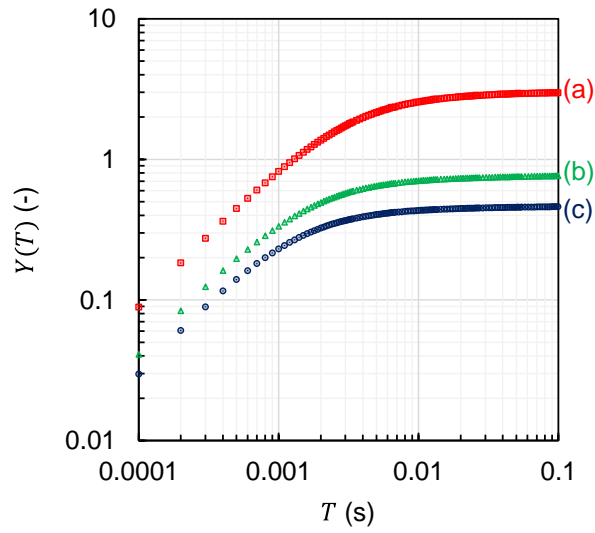
The measurement times of the reactor noises were approximately 139.4 min, 975.0 min, and 933.5 min for Cases (a)–(c), respectively. It should be noted that Case (a) was measured during operation; thus, the measurement time was shorter than those in the shutdown states of Cases (b) and (c). Using the recursive bunching method, the variations in $y_n(kT_0)$, $\frac{y_3(kT_0)}{(Y(kT_0))^2}$, and $\frac{y_4(kT_0)}{(Y(kT_0))^3}$ were evaluated for each measurement, where $T_0 = 10^{-4}$ (s) and $M = 1024$, and the total number of count data N_0 were approximately 8.363×10^7 , 5.850×10^8 , and 5.601×10^8 , respectively. To estimate the statistical errors of $y_n(kT_0)$, $\frac{y_3(kT_0)}{(Y(kT_0))^2}$, and $\frac{y_4(kT_0)}{(Y(kT_0))^3}$, the 95% bootstrap confidence intervals were also calculated using the moving block bootstrap method with $B = 1000$. Using the Feynman- α method [8,10,11] for the measured $Y(T)$, the prompt neutron decay constants α for Cases (a)–(c) were preliminarily estimated as 684.1 ± 3.5 , 1307.4 ± 4.0 , and 1618.5 ± 6.5 (1/s), respectively.

1 5.2. Experimental results

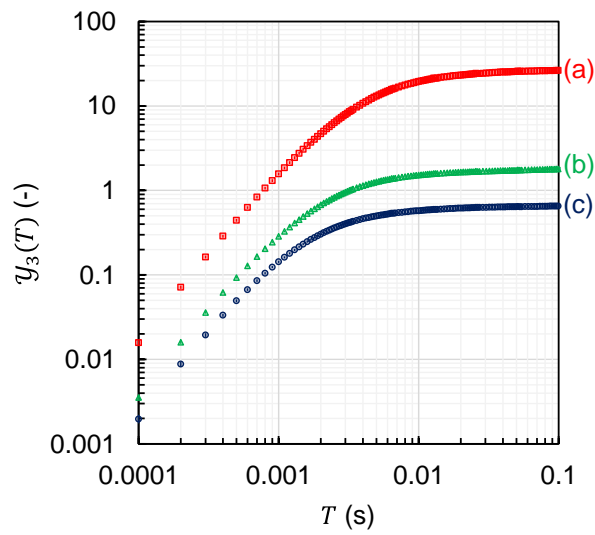
2 Figure 7 presents the variations in the second- to fourth-order neutron correlation factors
3 $Y(T)$, $\mathcal{Y}_3(T)$, and $\mathcal{Y}_4(T)$ with respect to the counting gate widths T . In Figure 7, the error
4 bars indicate the 95% bootstrap confidence intervals for $Y(T)$, $\mathcal{Y}_3(T)$, and $\mathcal{Y}_4(T)$. As can be
5 seen in Figure 7, it was confirmed that these values converge to the saturation values Y_∞ , $\mathcal{Y}_{3,\infty}$,
6 and $\mathcal{Y}_{4,\infty}$ as T becomes sufficiently large. As shown in Equations (43) and (45), the
7 magnitudes of Y_∞ , $\mathcal{Y}_{3,\infty}$, and $\mathcal{Y}_{4,\infty}$ are approximately inversely proportional to the second,
8 third, and fourth power of the subcriticality $-\rho$, respectively. Thus, these saturation values
9 decreased as $-\rho$ deepened.

10 Figure 8 presents the variation in the ratios of $\frac{\mathcal{Y}_3(T)}{(Y(T))^2}$ and $\frac{\mathcal{Y}_4(T)}{(Y(T))^3}$ with the 95% bootstrap
11 confidence intervals. In addition, Figure 9 presents the correlation matrices of the ratios
12 between different counting gate widths T . As can be seen from the correlation matrices, there
13 are strong correlations owing to the bunching method. These correlations decreased as the
14 difference between kT_0 and $k'T_0$ increased, and as the subcriticality deepened.

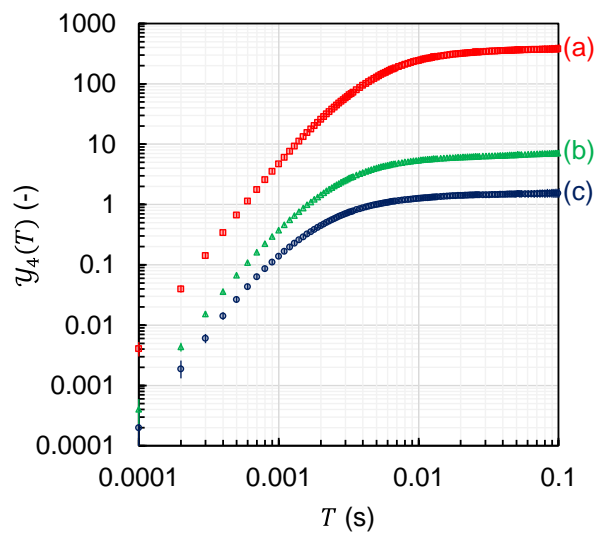
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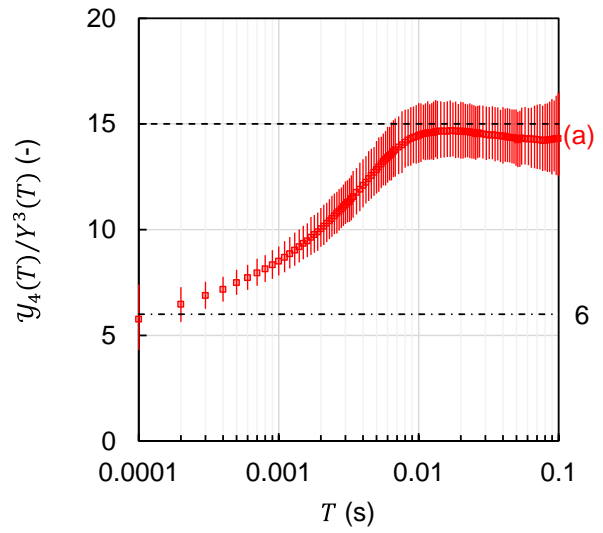
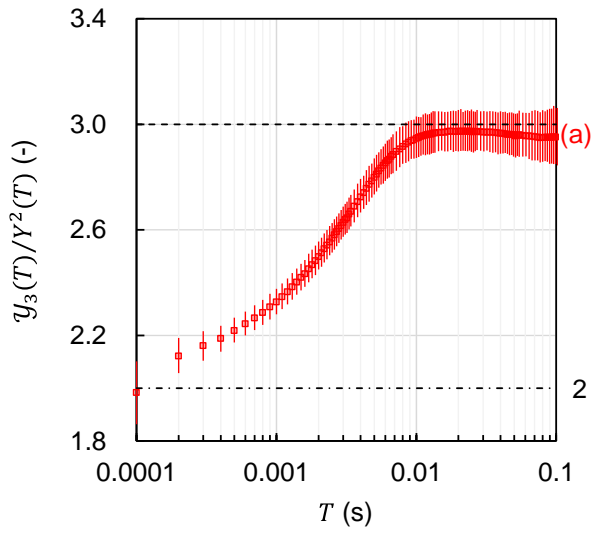
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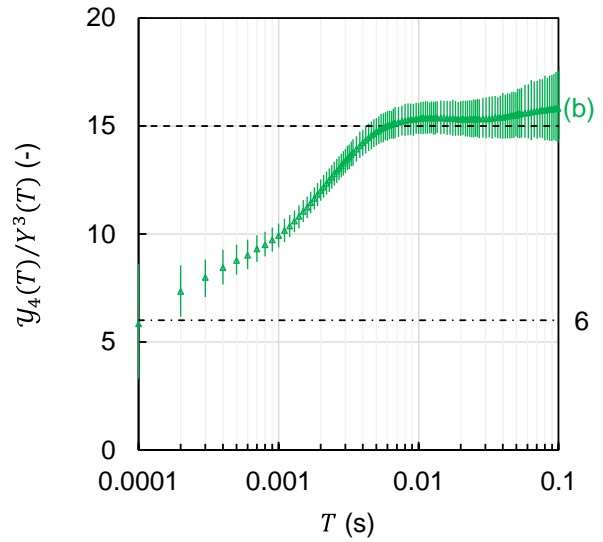
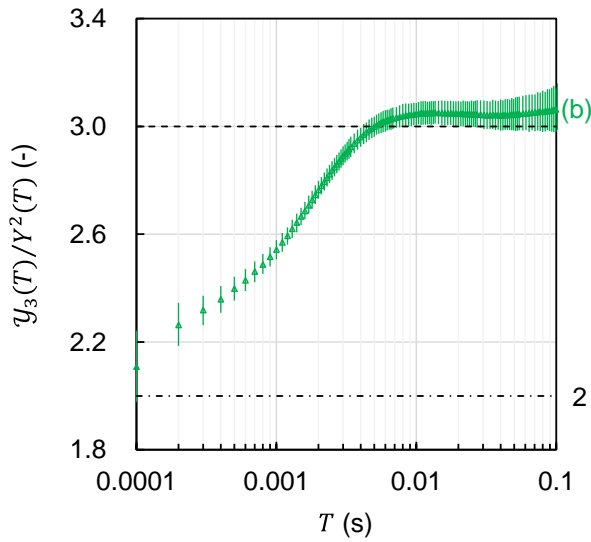
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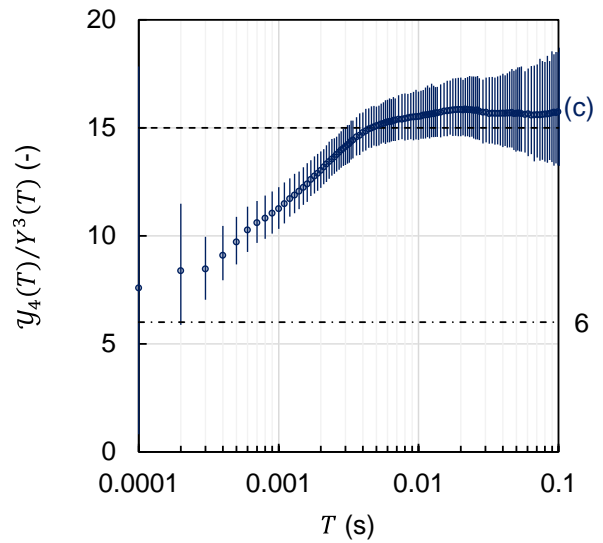
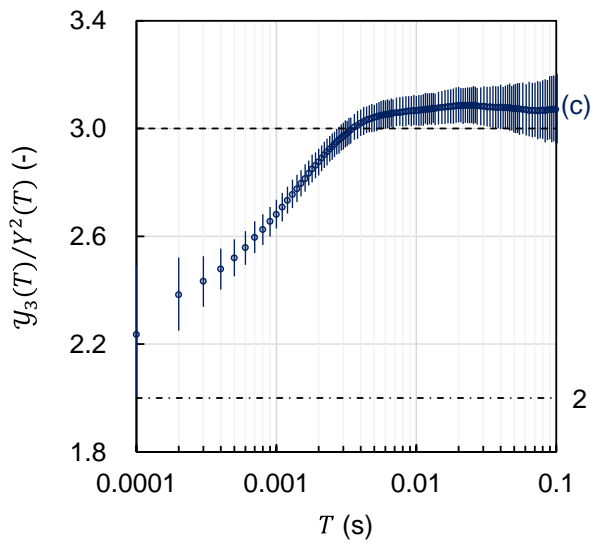
Figure 7 Experimental results of variations in $Y(T)$, $y_3(T)$, and $y_4(T)$



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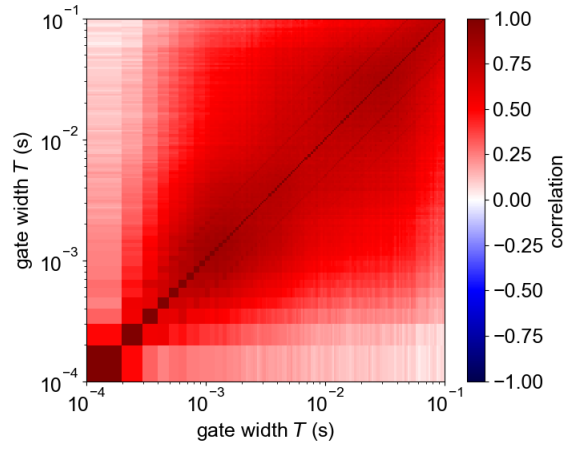
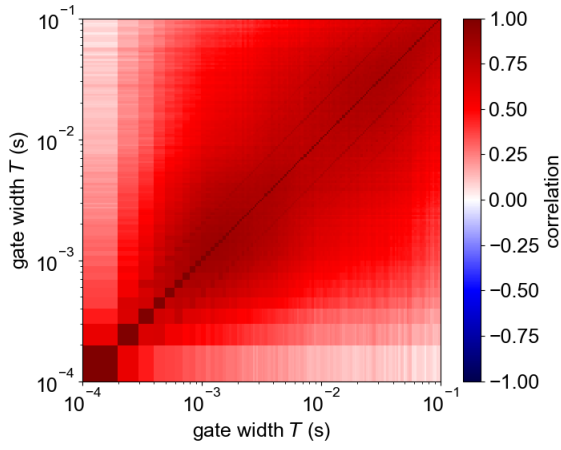
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Figure 8 Experimental results of variations in $\mathbf{y}_3(T)/(Y(T))^2$ and $\mathbf{y}_4(T)/(Y(T))^3$

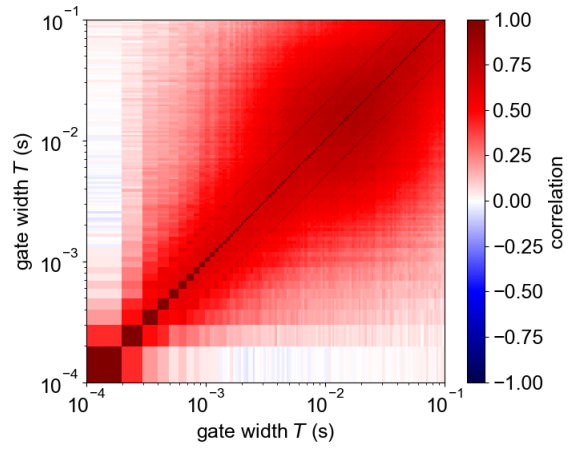
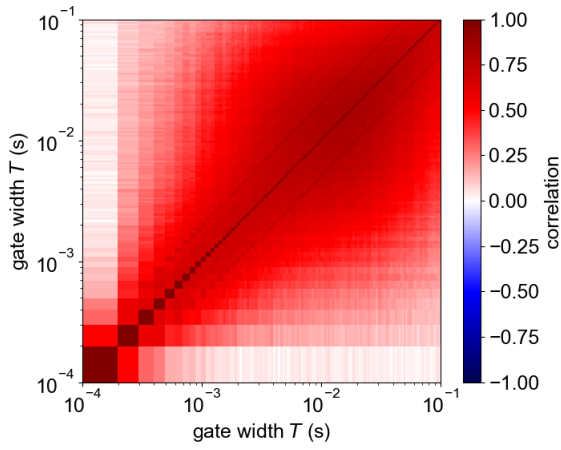


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Case (a) $y_3(T)/(Y(T))^2$

Case (a) $y_4(T)/(Y(T))^3$

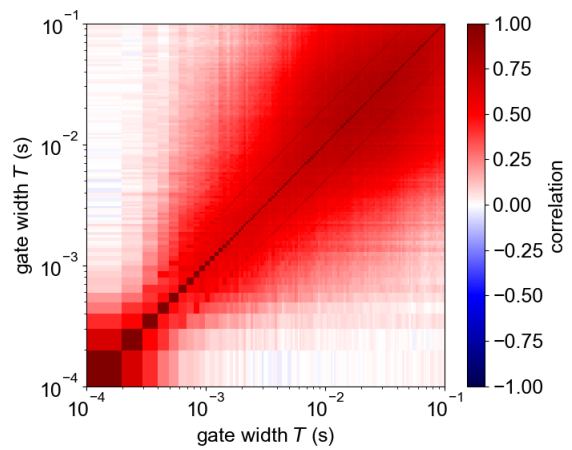
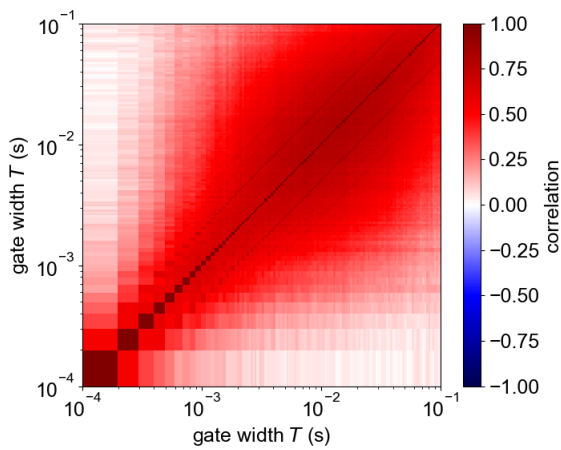


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Case (b) $y_3(T)/(Y(T))^2$

Case (b) $y_4(T)/(Y(T))^3$



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Case (c) $y_3(T)/(Y(T))^2$

Case (c) $y_4(T)/(Y(T))^3$

7

Figure 9 Experimental results of correlation matrices for $y_3(T)/(Y(T))^2$ and

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$y_4(T)/(Y(T))^3$

1

2 **5.3. Discussion**

3 As shown in Figure 8, $\frac{y_3(T)}{(Y(T))^2}$ and $\frac{y_4(T)}{(Y(T))^3}$ converge to the saturation values $\mathcal{Y}_{3,\infty}/Y_\infty^2$
4 and $\mathcal{Y}_{4,\infty}/Y_\infty^3$, as the gate width T (or the dimensionless quantity αT) becomes sufficiently
5 large. If T is approximately larger than $10/\alpha \approx 0.01$ (s), the ratios appear constant, although
6 there are statistical fluctuations. The 95% bootstrap confidence intervals of $\frac{y_3(T)}{(Y(T))^2}$ in Cases (a)
7 and (b) included the unique combination number ‘3,’ which was theoretically predicted by
8 Equation (50). In Case (c), it should be noted that there were slight differences between $\frac{y_3(T)}{(Y(T))^2}$
9 and ‘3’ when $0.01 < T < 0.03$ (s), whereas ‘3’ was found within the 95% bootstrap
10 confidence intervals when $T > 0.03$ (s). This observation confirms the relationships presented
11 in Equation (48). For example, when $T = 0.02$ (s), $\left(\frac{y_3(T)}{(Y(T))^2} - 3\right) \approx 0.08 \pm 0.03$ (95%
12 bootstrap confidence interval of [0.02, 0.15]), which approximately corresponds to the
13 magnitude of $-\rho \approx 0.07$ in Case (c). If the measurement time of the reactor noise is increased
14 by a factor of 4, the statistical significance of the difference between $\mathcal{Y}_{3,\infty}/Y_\infty^2$ and ‘3’ can be
15 detected, and $-\rho$ can be estimated more precisely using Equation (48). Similar to $\mathcal{Y}_{3,\infty}/Y_\infty^2$,
16 it was confirmed that the 95% bootstrap confidence intervals of the saturated $\frac{y_4(T)}{(Y(T))^3}$ in Cases
17 (a)–(c) included the unique combination number ‘15’ of Equation (51). Consequently,
18 Equations (50) and (51) were validated using the actual reactor noise measurement under the
19 subcritical core, where $-\rho < 0.1$.

20

21 From Figure 8, it can be seen that $\frac{y_3(T)}{(Y(T))^2}$ and $\frac{y_4(T)}{(Y(T))^3}$ tended toward to ‘2’ and ‘6’ as T
22 decreased to zero. This implies that the probability distribution of the neutron count $P(C, T)$
23 is sufficiently approximated by a negative binomial distribution under the condition of
24 $Y_\infty(\alpha T)^2 \ll 1$ and $\alpha T \ll 1$, as clarified by previous research [33,34,35].

$$P(C, T) \approx \frac{\Gamma(C+r)}{C! \Gamma(r)} \left(\frac{m}{r+m}\right)^C \left(\frac{r}{r+m}\right)^r, \quad (59)$$

1 where $\Gamma(x)$ represents the Gamma function; m and r are the population parameters of the
 2 negative binomial distribution for the reproduction of $P(C, T)$. The probability generating
 3 function for Equation (59) can then be expressed as follows:

$$G(Z, T) \approx \left(\frac{r}{r + (1-Z)m}\right)^r. \quad (60)$$

4 Using Equations (1) and (59), $\langle C(T) \rangle$ and the second-, third-, and fourth-order neutron
 5 correlation factors can be obtained as

$$\langle C(T) \rangle = \left. \frac{\partial G}{\partial Z} \right|_{Z=1} = m, \quad (61)$$

$$Y(T) = \left. \frac{1}{\langle C(T) \rangle} \frac{\partial^2}{\partial Z^2} \ln(G(Z, T)) \right|_{Z=1} = \frac{m}{r}, \quad (62)$$

$$y_3(T) = \left. \frac{1}{\langle C(T) \rangle} \frac{\partial^3}{\partial Z^3} \ln(G(Z, T)) \right|_{Z=1} = 2 \left(\frac{m}{r}\right)^2 = 2Y^2, \quad (63)$$

$$y_4(T) = \left. \frac{1}{\langle C(T) \rangle} \frac{\partial^4}{\partial Z^4} \ln(G(Z, T)) \right|_{Z=1} = 6 \left(\frac{m}{r}\right)^3 = 6Y^3. \quad (64)$$

6 From Equations (62)–(64), it was confirmed that $\frac{y_3(T)}{(Y(T))^2} \approx 2$ and $\frac{y_4(T)}{(Y(T))^3} \approx 6$ if the negative
 7 binomial distribution approximation is applicable to $P(C, T)$. In particular, the ratios of ‘2’ and
 8 ‘6’ are approximately half of these unique combination numbers, ‘3’ and ‘15.’ Thus, it was
 9 suggested that approximate orders of magnitudes for $y_3(T)$ and $y_4(T)$ can be estimated
 10 from the second and third power of $Y(T)$ over the whole range of T .

11

6. Future study prospects

As shown in Figure 1, the unique combination numbers of $\mathcal{Y}_{n,\infty}/Y_{\infty}^{n-1}$ near the critical state can be inferred by the heuristic enumeration method for $n \geq 5$. Based on the heuristic method for the complete (unordered) binary tree, as shown in Figure 1, the unique combination numbers $b_n \equiv \mathcal{Y}_{n,\infty}/Y_{\infty}^{n-1}$ can be deduced using the double factorial [36]:

$$b_n \equiv \frac{\mathcal{Y}_{n,\infty}}{Y_{\infty}^{n-1}} = \prod_{k=1}^{n-1} (2k-1) = (2n-3)!! \quad (65)$$

The sophisticated theoretical derivation for $\mathcal{Y}_{n,\infty}/Y_{\infty}^{n-1}$ for any system in the critical state is a topic of future work.

In Section 4, the virtual reactor noise measurement for the non-multiplication system with the ^{252}Cf source was verified with the aid of the Monte Carlo simulation. For the experimental validation, the actual measurement should be carried out, if possible. Hence, a high-efficiency neutron detection system is required to increase the magnitude of the neutron correlation factors $Y(T)$, $\mathcal{Y}_3(T)$, and $\mathcal{Y}_4(T)$.

In general, as the order of a neutron correlation factor $\mathcal{Y}_n(T)$ increases, the statistical error tends to increase. Hence, the experimental validation of the unique combination number for the significantly higher-order neutron correlation requires a longer measurement time of the reactor noise in a stable and unperturbed state such as the reactor shutdown state. From the experimental analysis conducted in this study, it can be concluded that $\mathcal{Y}_{3,\infty}/Y_{\infty}^2$ is relatively useful in comparison with $\mathcal{Y}_{4,\infty}/Y_{\infty}^3$ for the estimation of difference from the unique combination number, although the 95% confidence intervals of the experimental results were not sufficiently small. To reduce the statistical error of $\mathcal{Y}_{3,\infty}/Y_{\infty}^2$, the improvement of the detection efficiency and reduction of counting loss is necessary. Moreover, the measurement time should be increased depending on the subcriticality $-\rho$, because Equation (48) suggests that the difference between $\mathcal{Y}_{3,\infty}/Y_{\infty}^2$ and '3' increases as the subcriticality deepens.

1 In addition, the correlations of $\frac{y_3(T)}{(Y(T))^2}$ and $\frac{y_4(T)}{(Y(T))^3}$ between different counting gate
 2 widths T were significantly positive owing to the bunching method. Therefore, the saturation
 3 values in the fitting method or the averaging method should be carefully evaluated. In particular,
 4 if the correlations are neglected, the estimated standard errors for the saturation values may be
 5 underestimated. An advanced analysis methodology with consideration of the correlations
 6 should be conducted for the correct evaluation of the saturation values.

7

8 **7. Conclusion**

9 In this study, the fundamental physical properties of third- and fourth-order neutron
 10 correlation factors $y_3(T)$ and $y_4(T)$ in a source-driven subcritical system were theoretically
 11 derived. In particular, if the subcriticality $-\rho$ is approximately less than 0.1 (dk/k) = 10000
 12 (pcm), the saturation values of the ratios of $y_{3,\infty}/Y_\infty^2$ and $y_{4,\infty}/Y_\infty^3$ are almost equal to the
 13 unique combination numbers, ‘3’ and ‘15,’ independent of fissile materials and an external
 14 neutron source. The unique combination numbers, ‘3’ and ‘15,’ correspond to the total number
 15 of combinations for the trio- and quartet-detections, which have two and three two-forked
 16 branches, respectively, and are equal to the double factorial $(2n - 3)!!$.

17 On the other hand, in the case of the non-multiplication system, $y_{3,\infty}/Y_\infty^2$ and $y_{4,\infty}/Y_\infty^3$
 18 depend on the probability distribution of an external source $p_s(q)$. The ratios are significantly
 19 different from the unique combination numbers, ‘3’ and ‘15.’ Thus, the differences between
 20 $y_{3,\infty}/Y_\infty^2$ and ‘3’ and between $y_{4,\infty}/Y_\infty^3$ and ‘15,’ respectively, are useful information for the
 21 evaluation of whether the target system is in the near-critical state or not.

22 Using the moving block bootstrap method, the unique combination numbers, ‘3’ and ‘15,’
 23 for the third- and fourth-order neutron correlation factors were validated using the actual zero-
 24 power reactor noise measurements carried out at KUCA. The moving block bootstrap method
 25 enabled the estimation of the statistical errors of $\frac{y_3(T)}{(Y(T))^2}$ and $\frac{y_4(T)}{(Y(T))^3}$, in addition to the
 26 correlation matrices between different gate widths T owing to the bunching method. Based on

1 the experimental results, it was confirmed that approximate orders of magnitudes for $y_3(T)$
2 and $y_4(T)$ can be estimated using $y_3(T) \approx 3(Y(T))^2$ and $y_4(T) \approx 15(Y(T))^3$ over the
3 whole range of T .

4 Furthermore, the results of experimental analysis reveal that the ratio of $y_{3,\infty}/Y_\infty^2$ is
5 relatively useful in comparison with $y_{4,\infty}/Y_\infty^3$ for the estimation of the difference from the
6 unique combination number, although the 95% confidence intervals of the present experimental
7 results were not sufficiently small. The difference between $y_{3,\infty}/Y_\infty^2$ and '3' is useful
8 information for the statistical evaluation of whether the state is critical or subcritical, and for
9 the determination of the absolute value of the subcriticality $-\rho$. The focus of future work will
10 be directed toward the reduction of the statistical error of $y_{3,\infty}/Y_\infty^2$ and the development of an
11 advanced analysis methodology for $y_{3,\infty}/Y_\infty^2$ with consideration of the correlations owing to
12 the bunching method.

13

14

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