High Concentration Effects of Transmission Fluctuation Spectrometry with Temporal Correlation

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Abstract. Transmission fluctuation spectrometry (TFS) with temporal or spatial correlation is a recently-developed method for real-time, online/inline particle analysis in two-phase flows, whereby the particle size distribution (PSD) and particle concentration can be measured simultaneously. On the basis of a layer model and the assumption of geometrical ray propagation, an analytical expression of the transmission fluctuation spectrum can be obtained. It is found that the measurement matches the theory very well as long as the particle concentration is low enough. However, as the particle concentration increases, the measured transmission fluctuation spectrum deviates gradually from the theory, which is a result from the high concentration effects. The high concentration effects include those from particle overlapping and the monolayer structure. In this presentation, the high concentration effects are studied with numerical simulation. As a result, the empirical expressions for the correction of the high concentration effects are obtained.

1. INTRODUCTION
Real-time, online/inline particle size analysis plays an important role in many areas of industry. For instance, the size of pulverized coal used in the power plant is an important parameter that affects both combustion efficiency and atmospheric emissions of the plant. Particle size is particularly important in quality control applications and is necessary for control of critical aspects of a process. Till now, the development of the reliable technique for real-time, online/inline particle sizing remains a challenge. The theory of transmission fluctuation spectrometry (TFS) with correlation has recently been developed as a new method of particle size analysis, which can be used for real-time, online/inline applications. The theory is based on the layer model and the assumption of the geometrical ray propagation. An analytical expression of the transmission fluctuation correlation spectrum can be obtained. Two approaches to the TFS have been developed. One is the spatial correlation (transmission fluctuation spectrometry with spatial correlation, TFS-SC), in which two parallel narrow beams with a variable separation between each other are employed (Shen, et al., 2005a). However, the mechanical operation on the beam separation becomes challenging, especially for the measurements on small particles. The beams and the detectors should be able to move synchronously and fast enough in a micrometer scale. Furthermore, the beams should be consistent with each other both in the beam diameters and in propagating directions, which is very difficult to realize. So in real measurements, the temporal correlation is used with only one narrow beam (Shen et al., 2005b; Guo et al., 2005). This method is called as transmission fluctuation spectrometry with temporal correlation or auto-correlation (shortly TFS-TC or TFS-AC) (Yu et al., 2007a).
It is found that the measurement matches the theory very well as long as the particle concentration is low enough. However, if the particle concentration is high, the measurement deviates from the theory. So in this presentation we concentrate on the high concentration effects. Similarly to what we have done before, effects such as multiple scattering and dependent scattering can be excluded so that only the steric interactions between particles are accounted for (Shen et al., 2004, 2005c; Riebel et al., 2004). Steric interactions between particles are classified into those between particles within the same monolayer (i.e. the monolayer structure) and those between particles from different monolayers (known as particle overlapping), which cause different effects on the transmission fluctuation correlation spectrum. Theoretical and experimental studies show that, if the beam diameter is infinitely small compared to the particle size, the effect from particle overlapping can be excluded so that the transmission fluctuation spectrum is affected by the monolayer structure only (Shen et al., 2005a, b; Yu et al., 2007b). However, while the beam diameter is not very small compared to the particle size, effects from particle overlapping are much stronger than those from the monolayer structure (Guo et al., 2005; Yu et al., 2007a). Due to the mathematical difficulties, the high concentration is studied with simulation. As the result, the empirical expression is obtained for the correction of the high concentration effects on the TFS.

2. PRINCIPLE OF THE MEASUREMENTS

In the TFS-TC, the fluctuating transmission signal \( T(t) \) of a narrow beam is recorded in a very long period of sampling time \( t_s \). The expectancy of transmission product (ETP) is then calculated with a variable time difference (or the correlation time) \( \tau \):

\[
\text{ETP} = \lim_{t_s \to \infty} \frac{1}{t_s} \int_0^{t_s} T(t)T(t+\tau)dt
\]

(1)

By varying the correlation time, the ETP is obtained as a spectrum, which can be further solved to extract the information on particle size distribution and particle concentration simultaneously. When the particle concentration is not high, the ETP of a 3-dimensional particle system can be expressed as (Shen et al., 2005a):

\[
\text{ETP} = e \left\{ T(t) T(t+\tau) \right\} \approx \prod_{i-1}^N \left\{ T_{ML_i}(t) T_{ML_i}(t+\tau) \right\} \\
\approx e \left\{ T_{ML}(t) T_{ML}(t+\tau) \right\} = \left\{ T(t) T(t+\tau) \right\} = \left( \text{ETP}_{ML} \right)^N
\]

(2)

which means that the particle system can be taken as a pile of several monolayers. Therefore, we may start from a monolayer containing the mono-dispersed spherical particles. After a tedious deduction, the analytical solution of the ETP through a monolayer is obtained (Shen et al., 2007):

\[
\text{ETP}_{ML} = 1 - \beta \left[ 2 - \chi(\Delta, \Lambda) \right] + \beta^2 \left[ 1 + \varepsilon(\Delta, \Lambda, \beta) \right]
\]

(3)

Here, \( \beta \) is the monolayer density, which is related to the volume particle concentration \( C_v \) (i.e. \( \beta = PC_v \), \( P \geq 1.5 \) is the parameter of the suspension structure). The transition function \( \chi(\Delta, \Lambda) \) and \( \varepsilon(\Delta, \Lambda, \beta) \) are defined as:

\[
\chi(\Delta, \Lambda) = \int_0^{\infty} J_0(2u\Delta) \cdot \left[ \frac{2J_1(\Lambda \cdot u)}{\Lambda \cdot u} \right]^2 \cdot \frac{2J_2^2(u)}{u} \cdot du
\]

(4)

\[
\varepsilon(\Delta, \Lambda, \beta) = \int_0^{\infty} J_0(2u\Delta) \cdot \left[ \frac{2J_1(\Lambda \cdot u)}{\Lambda \cdot u} \right]^2 \cdot \frac{2J_2^2(u)}{u} \cdot F_{MS} \cdot du
\]

(5)

Here, \( \Delta = v \cdot \tau / \chi \) (\( v \) is the flow velocity) is called as the dimensionless correlation time, \( \Lambda = D / x \)
(D is the beam diameter) is the dimensionless beam diameter (or beam-to-particle diameter ratio), and x is the particle diameter. The factor \(2J_1(\Lambda u)/(\Lambda u)^2\) originates from spatial averaging over the cross section of the circular uniform beam, \(2J_1^2(u)/u\) describes the property of the spherical particle, and \(J_0(2u\Delta)\) is the effect of temporal correlation. The factor \(F_{MS}\) describes the monolayer structure, which is determined by the particle concentration and the particle interaction potential (Shen et al., 2003).

When the particle concentration is not high, the logarithm of the ETP of a 3-dimensional monodispersed particle system can be expressed as:

\[
\ln \text{ETP} = -\frac{1.5}{x} \Delta Z \cdot C_x \left[ 2 - \chi(\Delta, \Lambda) \right]
\]

(6)

Here, \(\Delta Z\) is the beam path in the measuring zone (i.e. the thickness of the particle system in the direction of ray propagation).

Eq.(6) can be extended to a poly-dispersed particle system, expressed as

\[
\ln \text{ETP} (\tau_j) = -\sum_i \frac{1.5}{\bar{x}_i} C_{i\gamma} (\bar{x}_i) \left[ 2 - \chi(\Delta_{i,j}, \Lambda_{i,j}, Z, \bar{x}_i) \right] \Delta Z
\]

(7)

\(\tau_j (j = 1, 2, \cdots, N)\) is the variable time difference for signal correlation. \(\bar{x}_i\), is the mean particle diameter in the \(i^{th}\) fraction of particle size \((i = 1, 2, \cdots, M)\) and \(C_{i\gamma} (\bar{x}_i)\) is the corresponding volume concentration. \(\Delta_{i,j} = \nu \tau_j / \bar{x}_i\) is the dimensionless correlation time.

Eq.(7) shows that the logarithm of the ETP is linear with respect to the particle concentration and the effects from the different particle size fractions superimpose linearly to form the logarithm of ETP. Therefore, particle size distribution and particle concentration can be retrieved by deconvoluting from the measurement of transmission fluctuations.

3. EFFECTS FROM THE MONOLAYER STRUCTURE

3.1 A Brief Discussion on the Monolayer Structure

The monolayer structure is determined by the particle concentration and the interaction potentials between particles. It is difficult to give an analytical expression exactly even for the simplest case of a system containing hard spherical particles, which corresponds to an interaction potential of infinity for interpenetrating particles and an interaction potential of zero for non-contacting particles. Only approximate solutions can be obtained. The monolayer structure is, in the theory of granularity of a monolayer, statistically characterized by means of the 2-dimensional particle pair correlation function (PPCF), \(g(r, \beta)\). Provided that the arrangement of spherical particles in the monolayer is isotropic and macroscopically homogeneous, the particle pair correlation function may be defined in terms of the radial distribution function:

\[
g(r, \beta) = \frac{dN(r)}{C_N \cdot 2\pi r \cdot dr}
\]

(8)

where \(dN(r)\) is the number of particles found in an annular zone of width \(dr\) at a distance \(r\) from a central particle, \(C_N = 4\beta / \pi x^2\) is the particle number concentration of the monolayer.

The PPCF can be numerically calculated with the Percus-Yevick approximation (Ripoll & Tejero, 1995; Shen et al., 2004). As shown in Fig.1, the PPCF varies as the monolayer density increases. In the limit of low particle concentration, the PPCF is
which means that an inter-penetration between hard particles in the single monolayer is prohibited and the particles can arrange randomly for distances of $r > x$. As the particle concentration increases, an increasingly oscillatory deviation from $g(r,0) = 1$ occurs in proximity to $r = x$. Both the amplitude and the reach of the oscillation grow with increasing concentration, indicating the gradual transition from disorder to low-range order and further to far-range order in the monolayer structure.

\[ g(r,0) = g(r,\beta)|_{\beta=0} = \begin{cases} 0 & \text{for } r/x \leq 1 \\ 1 & \text{else} \end{cases} \quad (9) \]

Fig. 1 The particle pair correlation function at different concentrations, $g(r,\beta)$.

### 3.2 Simulations on the Monolayer Structure

Numerical simulation on the monolayer is performed with monodisperse spherical particles contained in a monolayer of square shape. The extension of the monolayer is perpendicular to the radiation and is very large compared with the particle size so that the deviation from statistics could be somewhat tiny. The outline of the simulation was detailedly introduced by Shen et al., 2003. A new transition function of ETP$_{ML}$ is introduced, $\Xi_{ML} = \ln \{ETP_{ML}\}/\ln \{ET_{ML}\}$, whereby $ET_{ML} = 1 - \beta$ is the average transmission. Accordingly, the step height of the transition function is defined as:

\[ \Xi_{ML,SH} = \Xi_{ML}|_{\Delta \to \infty} - \Xi_{ML}|_{\Delta \to 0} = 2 - \Xi_{ML}|_{\Delta \to 0} \quad (10) \]

The relationship between $\Xi_{ML}$ and $\chi(\Delta, \Lambda)$ is found to be $\lim_{\beta \to 0} \Xi_{ML} = 2 - \chi$ in the limit of low particle concentration. As an example, Fig. 2 shows the theoretical and simulated results in which the beam-to-particle diameter ratio is $\Lambda = 1.0$. It can be found that, as the monolayer density increases, the step height of the transition function decreases and the transition function shifts to the range of smaller dimensionless correlation time gradually. The peak of the transition function $\Xi_{ML}$ appears close to $\Delta = 1$ with a slender fluctuant. Comparing Fig. 2a with Fig. 2b, it can be found that they show the similar change on the step height of the transition function and the dimensionless correlation time response. No more than, the change of the theoretical results is more obvious than that of the simulation results. The difference between the theoretical results and the simulated ones might be caused by both the approximation used in theoretical calculation of the PPCF and the deviation from statistics in the simulation due to the limited dimensions of the monolayer.
4. EFFECTS FROM PARTICLE OVERLAPPING

4.1 A Brief Discussion on Particle Overlapping

Shielding effects can be traced back to near-field phenomena. Considering the near field diffraction, the radiation incident upon the second monolayer is strongly dependent on particle arrangement in the first monolayer. Even at a very low concentration, when the particle arrangement in each monolayer is random, there are statistically a certain part of the particles in the second layer located in the shadows produced by those particles in the first layer. This interaction might be called as particle intersection or particle overlapping.

4.2 Simulations of Particle Overlapping

The high concentration effects due to particle overlapping can be quantified by the extinction of the irradiation \( E \), which is approximately a product of the monolayer density \( \beta \) and the number of layers \( N \) (Yu et al., 2007a).

\[
E = -\ln T = -\ln (1 - \beta)^N \approx \beta \cdot N
\]  

From this point of view, an increase of particle concentration is approximately equivalent to an increase of the layer number. However, the increase of particle concentration additionally leads to increasing effects from the monolayer structure. Therefore, in order to avoid any disturbance from the monolayer structure in the simulations, the particle concentration is assumed to be very low. Alternatively, this may be simulated either by assuming a large number of layers, each layer containing very few particles only, or by assuming a single, dense monolayer containing non-interacting particles, whereby particles are allowed to interpenetrate with each other. The dense monolayer containing \( N \) non-interacting particles is equivalent to a 3-dimensional particle suspension containing \( N \) layers, each layer containing one particle only and hence the monolayer density is

\[
\beta = \frac{\pi x^2}{4 \cdot l_1 l_2}
\]

As long as the dimensions of the monolayer, \( l_1 \) and \( l_2 \), are large enough, the monolayer density tends to zero so that the average transmission is expected to be:

\[
e(T) = \left(1 - \beta\right)^N \rightarrow e^{-\beta \cdot N}
\]  

A detailed description of the simulation on particle overlapping was given by Yu et al., 2007a so that only simulated results are presented here. As is given in Fig.3a, when the beam-to-particle diameter
ratio is $\Lambda = 0$, effects from particle overlapping are invisible. However, if the beam-to-particle diameter ratio equals 1, the step height of the transition function increases and the transition function shifts to the range of larger dimensionless correlation time gradually as the extinction increases. Similar effects are found for different values of $\Lambda$, which means the effects from particle overlapping have the same tendency and are dependent on the beam-to-particle diameter ratio $\Lambda$.

![Simulated results of particle overlapping.](image)

**Fig. 3** Simulated results of particle overlapping.

### 5. THE HIGH CONCENTRATION EFFECTS

Usually, the particle concentration is not low so that particle overlapping and the monolayer structure exist simultaneously, leading the high concentration effects to be more complicated. Fig. 4a shows the dependence of the transition functions on the extinction, in which the number of layers is $N = 40$ and the beam-to-particle diameter ratio is $\Lambda = 1.0$. It can be found that, as the extinction increases, the step height of the transition function increases and the transition function shifts to the range of larger dimensionless correlation time gradually. And the simulated curves are below the theoretical curve in the range of the dimensionless correlation time $\Delta < 1$. This means that the transmission fluctuation spectrum is mainly affected by particle overlapping. As a comparison, the transition functions for the number of layers $N = 5$ and the beam-to-particle diameter ratio $\Lambda = 1.0$ are simulated, as shown in Fig. 4b. It can be found that, as the extinction increases, the step height of the transition function decreases and the transition function shifts to the range of smaller dimensionless correlation time gradually. All the simulated curves are over the theoretical curve in the range of the dimensionless correlation time $\Delta < 1$. This is because that, in the former case, the number of layers is large and the monolayer density (or the particle concentration) is not very high so that effects from the monolayer structure are weaker than those from particle overlapping. However, in the latter case, the layer number is small and the particle concentration is high so that effects from monolayer structure are much stronger than those from particle overlapping.
6. EMPIRICAL CORRECTION FROM MEASUREMENTS

6.1 Correction of the High Concentration Effects on the Spectrum Response

In order to see the high concentration effects on the response of the transmission fluctuation spectrum, the transition function is introduced into a normalized transition function, separating out the change of the step height. The normalized transition function \( \Xi_N(\Delta) \) is defined as:

\[
\Xi_N(\Delta) = \frac{\Xi(\Delta) - \Xi(\Delta \to 0)}{\Xi(\Delta \to \infty) - \Xi(\Delta \to 0)} + 1
\]

(14)

When \( 0 < \Lambda \leq 4 \), the high concentration effects on the normalized transition function can be empirically expressed as the deviation from theoretical normalized transition function in low concentration limit, defined as:

\[
\Delta \Xi_N(\Delta) = \left[ \Xi_N(\Delta) - \Xi_N(\Delta, C_V \to 0) \cdot e^{-(\Lambda+0.546)\Delta} \cdot e^{\left[\frac{(\Delta-0.546)^2}{4} + \frac{\Lambda}{10}\right] \Delta} \right]
\]

(15)

Here, \( \Xi_N(\Delta, C_V \to 0) \) is the theoretical normalized transition function in low concentration limit.

In order to take a deeper look, the deviation of the normalized transition function is factorized into a product of the Module \( M \) and the normalized deviation \( \zeta(\Delta) \). \( M \) is the average of the maximum of the deviation of the normalized transition function \( \Delta \Xi_N(\Delta) \):

\[
\Delta \Xi_N(\Delta) = M \cdot \zeta(\Delta)
\]

(16)

The normalized deviation \( \zeta(\Delta) \) depends on the dimensionless beam diameter, which can be described with a translation of the dimensionless correlation time of:

\[
\Delta' = \Delta \cdot \left[ \frac{1 + 0.285 \cdot (\Lambda + 0.45) \cdot e^{-0.546 \cdot \Delta}}{1 + 0.546 \cdot (\Lambda + 0.1)} \right]
\]

(17)

and the distribution of the normalized deviation might be described with an empirical expression:

\[
\zeta(\Delta') = \left( \frac{e}{a} \right) \cdot e^a \cdot e^{-e^a} \cdot \frac{(\Delta')^c}{b}
\]

(18)

The parameters are \( a = 1.31843 \), \( b = 0.162711 \) and \( c = 0.860572 \).
Fig. 5 shows the dependence of $M$ on $\Lambda$, with different values of $E$ and $N$. As shown in Fig. 5a, when $N$ keeps unchanged (e.g. $N = 40$) and $E$ is 0.05, 2.0 and 4.0 respectively, $M$ increases along with the increase of $\Lambda$. These three curves are approximately consistent with each other. Similarly, when $E$ is unchanged (e.g. $E = 0.05$) and $N$ is 5, 10, 40 respectively, $M$ increases along with the increase of $\Lambda$ (see Fig. 5b). Therefore, $M$ is related to the beam-to-particle diameter ratio $\Lambda$ only and almost independent of $E$ and $N$. This is quite different from what we obtained before in TFS with signal averaging or filtering (Xu et al., 2006). The approximate expression is obtained

$$M = M(\Lambda) = 0.028\Lambda^2 + 0.061\Lambda + 0.087$$

From the simulation above, we may conclude that effects from particle overlapping are closely related to the extinction $E$, the layer number $N$ and the beam-to-particle diameter ratio $\Lambda$.

6.2 Correction of the High Concentration Effects on the Step Height

In order to correct the high concentration effects on the step height of the transition function, another function $K$ is introduced, which is defined as $K = \Xi(\Delta = 0)$. The empirical expression is found from the simulation:

$$K = K(E) = f \cdot E^2 + g \cdot E + h$$

Here, $f, g, h$ are found to depend on the beam-to-particle diameter ratio $\Lambda$:

$$f = -0.002\Lambda$$

$$g = 0.012\Lambda^2 - 0.044\Lambda$$

$$h = -0.115\Lambda^2 + 0.650\Lambda + 1$$

One special case is that the particle is much larger than the beam diameter, i.e. the beam-to-particle diameter ratio is $\Lambda = 0$. In this case, no matter the particle system is thin or thick, the function is found to be $K|_{\Lambda = 0} = 1$, which was proved by both the simulations of $N = 5, 10, 20, 30, 40$ and the theory (Shen et al., 2005a, b; Yu et al., 2007b). Since the beam-to-particle diameter ratio is equal to 0, the step height of the transition function keeps unchanged. This implies that, if the beam diameter is infinitely small compared to the particle size, effects from particle overlapping are so small that the transmission fluctuation spectrum is affected by the monolayer structure only. It should be pointed out that another special case occurs when the number of layers is very small, wherein the high concentration effects arise mainly from the monolayer structure. As it was given in the simulation (see Fig. 2), in this case the step height decreases when the particle concentration increases. The variation of
the step height caused by the monolayer structure depends closely on the beam-to-particle diameter ratio. However, this situation is not very important because in real measurements the particle system generally includes a lot of layers.

Fig.6 shows the simulated effects on the step height from particle overlapping (i.e. effects from the monolayer structure are excluded, please refer to sec.4.2), wherein the values calculated with the empirical expression are plotted in lines and the simulated results are given with dots. The match between the empirical expression and the simulation is satisfactory.

For particle systems which contain both the monolayer structure and particle overlapping, simulations are given in Figs.7 & 8. All the values calculated with the empirical expression are plotted in lines and the simulated results are given with dots. In Fig.7, the number of layers is \( N = 20 \) and the beam-to-particle diameter ratio is 0.1, 0.5, 1.0, 2.0 and 3.16 respectively.

In Fig.8, the beam-to-particle diameter ratio is \( \Lambda = 1 \) and the number of layers is \( N = 5, 10, 20, 30, 40 \) respectively. It can be found that all these curves except for \( N = 5 \) decreases along with the increase of the extinction. This is because that, when \( N = 5 \), the effects from the
monolayer structure are stronger than those from particle overlapping. In all the cases plotted in Figs. 6-8, the empirical expression for the step height can match the simulated results very well.

![Graph showing the dependence of K on the extinct E](image)

**Fig.8 Dependence of K on the extinct E (Λ = 1).**

### 7. CONCLUSIONS

In this presentation, the high concentration effects on the transmission fluctuation spectrum with temporal correlation are studied. It is found that, in usual cases, these effects arise from both particle overlapping and the monolayer structure. The increase of particle concentration enhances the effects from both the monolayer structure and particle overlapping, while the increase of the layer number enhances the effects from particle overlapping only. The monolayer structure shifts the transmission fluctuation spectrum to the range of lower correlation time and reduces the step height of the transition function. On the contrary, particle overlapping shifts the transmission fluctuation spectrum to the range of higher correlation time and increases the step height of the transition function. Therefore, in real measurements, there is a balance between the high concentration effects from the monolayer structure and those from particle overlapping. Usually, the particle concentration is not too high and the particle system is thick enough to contain a lot of layers. Thus, effects from particle overlapping are much stronger than those from the monolayer structure.

The simulated results show that the high concentration effects are closely related to the beam-to-particle diameter ratio, the number of layers and the particle concentration (or the extinction). Analyzing the simulated results, the empirical expressions are achieved which indicate both the variation of the temporal response of spectrum and the variation of the step height of the transition function. The empirical expressions may be hopefully used to correct the high concentration effects in the inverse problem to extract particle size distribution and particle concentration.

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### NOMENCLATURE

| Symbol | Description     |
|--------|-----------------|
| $C_N$  | particle number concentration |
| $C_V$  | particle volume concentration |
| $D$    | beam diameter [m] |
| $e(\cdot)$ | expectancy of |
The average transmission

The expectancy of transmission product, \( e(T_1 T_2) \)

Factor of the monolayer structure

Particle pair correlation function

Bessel function

the function with parameters of \( f, g \) and \( h \)

Length of the edges of the monolayer in simulations \([m]\)

Module of the deviation (with parameters of \( k_1, k_2 \) and \( k_3 \))

Number of monolayers

Monolayer structural parameter \( P \geq 1.5 \)

The particle size distribution

Transmission fluctuation signal

Transmission fluctuation spectrometry

Transmission fluctuation spectrometry with auto-correlation

Transmission fluctuation spectrometry with spatial correlation

Transmission fluctuation spectrometry with temporal correlation

Sampling time \([s]\)

Integration variable

Particle flow velocity \([m/s]\)

Particle diameter \([m]\)

The mean particle diameter in the \( i^{th} \) fraction of particle size \([m]\)

Monolayer density

Dimensionless correlation time, \( \Delta = \nu \cdot \tau / x \)

The beam path in the measuring zone \([m]\)

Deviation of the transition function, of the normalized transition function

Distribution of the normalized deviation (with parameters of \( a, b \) and \( c \))

Dimensionless spatial averaging parameter / beam-to-particle diameter ratio \( \Lambda = D / x \)

Transition function of ETP, normalized transition function

Step height of the transition function, defined as the difference between the higher level and the lower level of the transition function, i.e. \( \Xi_{SH} = 2 - \Xi(\Delta \to 0) \)

Correlation time

Transition function

Variables of the number
ML  monolayer  
N normalized  
SH step height, defined as the difference between the higher level and the lower level of the transition function  
V volume

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