# Collision rates in chaotic flows: Dilute suspensions

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We have computed collision rates between passive marker particles subject to the blinking vortex flow, a chaotic flow. The case of low number densities is considered, which is relevant for the later stages of the aggregation processes. The theory pertaining to this case is first reviewed. The computational results indicate that the time-averaged collision frequency achieves an asymptotic value  $(\langle f \rangle_{\infty})$  for all number densities that is nearly independent of the initial positions of the particles. The dependence of  $\langle f \rangle_{\infty}$  on the system parameters was found to be similar to the predictions of the gradient theory of Levich [*Physico-Chemical Hydrodynamics* (Prentice Hall, Englewood Cliffs, NJ, 1962), Sec. 40]; the magnitude of the collision rate was significantly smaller. The deviation, attributed to the structure in the flow, decreases rapidly with increasing number density. The results imply validity of the gradient theory for low number densities for chaotic flows.

### INTRODUCTION

Aggregation processes can be considered to comprise two basic components: "collision" between two particles followed by a "reaction." In the limit of an instantaneous reaction step, every collision results in aggregation, and the particle kinematics determine the overall rate of the process. Collision rates thus provide the basic information for determining aggregation rates and have been obtained for a number of systems.<sup>1-5</sup> Flow kinematics and particle kinematics coincide when Brownian forces, particle-particle interactions, and inertial forces are neglected. Particles then act as passive nondiffusive tracers of flow, and the aggregation rate is determined by how frequently pairs of trajectories approach within the collision diameter. For a simple shear flow, and spherical particles, the collision frequency is given by the classical gradient theory of Levich.<sup>5</sup> We examine the applicability of the theory to the case of chaotic flows here.

Muzzio and Ottino<sup>6</sup> recently studied the pairwise coagulation in the blinking vortex flow,<sup>7,8</sup> a chaotic flow, for a large number of particles. Upon collision, one of the particles was assumed to disappear while the "order" of the second was increased. Consequently, there was a tendency for local reduction in the number density of particles in regions of higher shear rate. When the rate of mixing (characterized by a parameter  $N_{mix}$ ) was sufficiently large, the number-density fluctuations were smoothed out and the computational results matched closely with those predicted by the gradient theory. In the case of regularly spaced drops on square grid, coagulation occurred only in particular regions of the flow, indicating the importance of location of the drops.

We have computed the collision rates in the limit of very dilute suspensions for the blinking vortex flow. The local depletion of particles observed by Muzzio and Ottino<sup>6</sup> does not occur since the particles are assumed to remain unaffected by collisions. We have considered the effect of the parameters of the system on the dilute suspension collision rate, and the crossover that occurs

while going from a dilute to a nondilute suspension. The latter by definition corresponds to the number density when the gradient theory is valid. The results are useful for understanding the behavior at later stages of aggregation processes in chaotic flows, and as a probe of the statistical properties of the flow.

In the following sections we first define the blinking vortex flow, and review the theory for collision rates in laminar flows. The computational results and discussion are then presented.

#### **BLINKING VORTEX FLOW**

The blinking vortex flow is a kinematically defined flow comprising two corotating point vortices separated by a fixed distance 2a. Each vortex acts alternately for a time T with a circulation  $\Gamma$ , and the dimensionless parameter governing the flow is the flow strength

$$\mu = \Gamma T / (2\pi a^2) . \tag{1}$$

The system has been analyzed in detail<sup>7,8</sup> and we briefly enumerate some of the pertinent results below. At low flow strengths ( $\mu \approx 0.01$ ) the flow is regular on a large scale, almost everywhere. With increasing flow strength macroscopic regions of chaotic flow [i.e., of length scale O(a)] appear, and at  $\mu_c \approx 0.36$  there is a transition to global chaos when three unconnected regions merge, to form a single one. The chaotic region formed is bounded by a Kolmogorov-Arnold-Moser (KAM) curve, and chains of small islands are evident near the boundary. At flow strengths slightly greater than  $\mu_c$  a cantorus encircling each vortex acts as a partial barrier to the flow. The Liapunov exponents calculated for the flow are O(1) and are nearly independent of the initial position.

The area of the chaotic region has a complex shape, and was obtained computationally. The boundary of the chaotic region was found by repeatedly mapping a particle and noting its maximum radial distance from a reference point at different discretized angular positions. The area of the chaotic region was then found by summing

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FIG. 1. Boundary of the chaotic region used to calculate the area with the corresponding Poincaré section for the blinking vortex flow. (a)  $\mu = 0.4$ , (b)  $\mu = 0.75$ , and (c)  $\mu = 0.8$ .

the areas of the sectors. As is evident from Fig. 1, the boundary used to calculate the area matches closely with that of the corresponding Poincaré sections. The area of the chaotic region has a complicated dependence on flow strength (Fig. 2): near  $\mu = 0.75$  there is a sharp decrease in area, and in some ranges of  $\mu$  computation of the area is difficult due to the presence of large islands.<sup>9</sup>



FIG. 2. Graph of the area of chaotic region (A) vs the flow strength  $(\mu)$  for blinking vortex flow. A is not calculated for  $\mu \in (1.0-1.5)$  and  $\mu > 2.5$  because of large islands in the chaotic region.

#### THEORY

We consider pairwise collisions between n circular particles of diameter d on a plane. The particles move as passive tracers in the flow so that

$$\mathbf{r}_{i} = \boldsymbol{\phi}_{i}(\mathbf{r}_{i}^{0}), \quad \mathbf{r}_{i}^{0} = \boldsymbol{\phi}_{0}(\mathbf{r}_{i}^{0}) , \qquad (2)$$

where  $\mathbf{r}_i$  is the position of the center of the particle *i* at time *t*, and  $\boldsymbol{\phi}_t$  is the flow.  $\boldsymbol{\phi}_t$  acts on an area *A*, with a normalized Lesbegue measure  $d\alpha = dA/A$ , which is preserved by the flow. The triplet  $(\boldsymbol{\phi}_t, \alpha, A)$  defines a dynamical system.<sup>10</sup>

The average collision frequency in this case is given by

$$\langle f \rangle_{\infty} = \lim_{t \to \infty} (1/4t) \sum_{i=1}^{n} \sum_{j=1}^{n} \int_{c_{ij}} \delta(S_{ij}) dS_{ij} ,$$
 (3)

where  $\delta(x)$  is the Dirac delta function, and

$$S_{ij}(t) = ||\mathbf{r}_i - \mathbf{r}_j| - d|$$
(4)

is the shortest distance between the perimeter of particles i and j. The integral is over the trajectories of each pair of particles i and j, and the factor 4 appears since every collision involves two particles and two zeros of the distance function  $S_{ij}(t)$ . Equation (3) can be written more conveniently as

$$\langle f \rangle_{\infty} = \lim_{t \to \infty} (1/4t) \sum_{i=1}^{n} \sum_{j=1}^{n} \int_{0}^{t} \delta(S_{ij}) \frac{dS_{ij}}{d\tau} d\tau$$
 (5a)

$$= \lim_{t \to \infty} (1/4t) \sum_{i=1}^{n} \sum_{j=1}^{n} \int_{0}^{t} g(\mathbf{r}_{i}, \mathbf{r}_{j}) d\tau .$$
 (5b)

We examine below the conditions under which the above time average can be replaced by a space average, which is much simpler to evaluate.

Defining a new phase space  $B_{ij}$  with elements  $\mathbf{x}_{ij} = (\mathbf{r}_i, \mathbf{r}_j)$ , and a flow  $\boldsymbol{\psi}_t(\mathbf{x}_{ij}^0) = (\boldsymbol{\phi}_t(\mathbf{r}_1^0), \boldsymbol{\phi}_t(\mathbf{r}_j^0))$ , we obtain the collision frequency as

$$\langle f \rangle_{\infty} = \lim_{t \to \infty} (1/4t) \sum_{i=1}^{n} \sum_{j=1}^{n} \int_{0}^{t} g(\mathbf{x}_{ij}) d\tau .$$
 (6)

The normalized measure in this case is  $d\beta_{ij} = dA_i dA_j / A^2$  and is preserved by the flow.  $\psi_i$  as defined above is said to be the direct product of

 $\boldsymbol{\psi}_t$  as defined above is said to be the direct product of  $\boldsymbol{\phi}_t$  with itself, and the following relations hold between them:<sup>10</sup> (i) if  $\boldsymbol{\psi}_t$  is ergodic then  $\boldsymbol{\phi}_t$  is ergodic; (ii) if  $\boldsymbol{\psi}_t$  is weak mixing then  $\boldsymbol{\phi}_t$  is weak mixing; (iii) if  $\boldsymbol{\psi}_t$  is mixing then  $\boldsymbol{\phi}_t$  is mixing. The converse of the above statements is true for (ii) and (iii) but not for (i). Thus  $\boldsymbol{\phi}_t$  must at least be weak mixing for  $\boldsymbol{\psi}_t$  to be ergodic. In this case

$$\langle f \rangle_{\infty} = f_A = \frac{1}{4} \sum_{i=1}^n \sum_{j=1}^n \int_{\beta_{ij}} \delta(S_{ij}) \frac{dS_{ij}}{d\tau} d\beta_{ij} , \qquad (7)$$

where  $f_A$  denotes the space average. Since every pair of trajectories is identical we obtain

$$f_A = \frac{1}{4}\rho^2 \int_A \int_A \delta(S_{ij}) \frac{dS_{ij}}{d\tau} dA_i dA_j , \qquad (8)$$

where  $\rho = n/A$  is the number density. (Note that the

number density appears outside the integral.) From the definition of  $S_{ij}$  we get

$$\frac{dS_{ij}}{dt} = \frac{|(\dot{\mathbf{r}}_i - \dot{\mathbf{r}}_j) \cdot (\mathbf{r}_i - \mathbf{r}_j)|}{|\mathbf{r}_i - \mathbf{r}_j|} = |[\mathbf{v}(\mathbf{r}_i) - \mathbf{v}(\mathbf{r}_j)] \cdot \hat{\mathbf{r}}_{ij}| , \qquad (9)$$

where **v** is the imposed velocity and  $\hat{\mathbf{r}}_{ij}$  is a unit vector along the line joining the center of particle *j* to particle *i*. By defining  $\mathbf{p} = (\mathbf{r} - \mathbf{r}_i)$  and  $\mathbf{p}_i = (\mathbf{r}_i - \mathbf{r}_i)$  we get

$$f_{A} = (\rho^{2}/4) \int_{A} dA'_{i} \int_{A} p_{j} \delta(|p_{j} - d|) \\ \times |[\mathbf{v}(\mathbf{0}) - \mathbf{v}(\mathbf{p}_{j})] \cdot \hat{\mathbf{r}}_{ij} | d\theta_{j} dp ,$$
(10)

where  $p_j = |\mathbf{p}_j|$  and  $\theta_j$  is the angle between  $\hat{\mathbf{r}}_{ij}$  and a reference line. Upon simplification we finally obtain

$$f_{A} = (\rho^{2}d/4) \int_{A} dA_{i} \int_{0}^{2\pi} |[\mathbf{v}(\mathbf{r}_{i}) - \mathbf{v}(\mathbf{r}_{i} + \mathbf{\hat{r}}d)] \cdot \mathbf{\hat{r}}(\theta)| d\theta ,$$
(11)

where  $\hat{\mathbf{r}}(\theta)$  is the unit outward normal vector to the perimeter of particle *i*, and  $\theta$  is the angle between  $\hat{\mathbf{r}}$  and a reference line.

Ergodicity of  $\psi_t$ , however, is not required for the ergodicity of the system (i.e., for  $\langle f \rangle_{\infty} = f_A$ ) if the system size is sufficiently large. In the limit  $n \to \infty$ , for fixed number density, the collision frequency is time independent if the particles are uniformly distributed initially  $(\nabla \rho|_{t=0}=0)$  or if  $\mathbf{v} \cdot \nabla \rho = 0$ . In this case the collision frequency is

$$\langle f \rangle_{\infty} = f_{A\rho} = \lim_{n \to \infty} \frac{1}{4} \sum_{i=1}^{n} \sum_{j=1}^{n} g(\mathbf{r}_{i}, \mathbf{r}_{j}) , \qquad (12)$$

Replacing the sum by an integral over the probability distribution of  $(\mathbf{r}_i, \mathbf{r}_j)$  we obtain

$$f_{A\rho} = \lim_{n \to \infty} \frac{1}{4} \int_{A} \int_{A} \rho(\mathbf{r}_{i}) \rho(\mathbf{r}_{j}) g(\mathbf{r}_{i}, \mathbf{r}_{j}) dA_{i} dA_{j} , \quad (13)$$

which upon simplification becomes

$$f_{A\rho} = \lim_{n \to \infty} \frac{1}{4} \int_{A} dA_{i} \rho(\mathbf{r}_{i}) \int_{0}^{2\pi} d\theta \rho(\mathbf{r}_{i} + \hat{\mathbf{r}}d) \times |[\mathbf{v}(\mathbf{r}_{i}) - \mathbf{v}(\mathbf{r}_{i} + \hat{\mathbf{r}}d)] \cdot \hat{\mathbf{r}}(\theta)| .$$
(14)

In the case of uniform density we get

$$f_{A} = (\rho^{2}d/4) \int_{A} dA_{i} \int_{0}^{2\pi} |[\mathbf{v}(\mathbf{r}_{i}) - \mathbf{v}(\mathbf{r}_{i} + \hat{\mathbf{r}}d)] \cdot \hat{\mathbf{r}}(\theta)| d\theta ,$$
(15)

which is identical to Eq. (11).

Further simplification of Eq. (15) is possible for special cases. For sufficiently small particles  $(d |\nabla \nabla \mathbf{v}| / |\nabla \mathbf{v}| \ll 1)$ 

$$f_{A} = (\rho^{2} d^{2} / 4) \int_{A} dA \int_{0}^{2\pi} d\theta | \mathbf{D} \cdot \hat{\mathbf{r}} \cdot \hat{\mathbf{r}} | , \qquad (16)$$

where D is the symmetric part of the velocity gradient tensor. In the case of homogeneous flows for any particle size Eq. (15) reduces to

$$f_{A} = (\rho^{2} d^{2} / 4) A \int_{0}^{2\pi} d\theta |\mathbf{D} \cdot \hat{\mathbf{r}} \cdot \hat{\mathbf{r}}| .$$
(17)

Finally, for simple shear flow we obtain

$$f_A = \rho^2 d^2 A \dot{\gamma} / 2$$
, (18)

using  $D = (\dot{\gamma}/2)(\hat{\mathbf{e}}_1\hat{\mathbf{e}}_2 + \hat{\mathbf{e}}_2\hat{\mathbf{e}}_1)$ . Equation (18) is the twodimensional (2D) analog of the expression proposed by Levich<sup>5</sup> for simple shear flow.

In the case of systems of finite size, and when the imposed flow is regular, the collision frequency  $(\langle f \rangle_{\infty})$  will deviate from the theoretically predicted value  $(f_A)$ . We estimate this deviation for the case of a simple shear flow  $(v_x = \dot{\gamma}y, v_y = 0)$ . Consider the collision frequency of *n* circular particles of diameter *d* uniformly distributed on a square of side *L* with the edge x = 0 joined to x = L. The collisions experienced by particle *i* are given by

$$\left( \dot{j}_{i} \right)_{\infty} = \dot{\gamma} d^{2} n_{i} / A_{i} , \qquad (19)$$

where  $n_i$  are the number of particles uniformly distributed in a strip

$$A_i = \{x, y : y \in (y_i - d, y_i + d), x \in (0, L)\},$$
(20)

and  $y_i$  is the position of particle *i*. In deriving Eq. (19), the fluctuations in  $\langle \mathcal{F}_i \rangle_{\infty}$  due to the fluctuations in number density inside the strip are neglected. The collision frequency is then

$$\langle f \rangle_{\infty} = \frac{1}{2} \sum_{i=1}^{n} \dot{\gamma} d^2 n_i / A_i , \qquad (21)$$

and the mean value  $(f_A)$  and the standard deviation of  $\langle f \rangle_{\infty}$  are obtained by taking an average over the probability distribution of  $n_i$  given by

$$P(n_i) = \frac{n!}{n!(n-n_i)!} q^{n_i} (1-q)^{n-n_i} , \qquad (22)$$

where  $q = A_i / A$ . Thus

$$\langle f \rangle_{\infty} = f_A \left[ 1 \pm \frac{1}{\sqrt{n}} (L/2d - 1)^{1/2} \right]$$
 (23a)

$$\approx f_A \left[ 1 \pm \left[ \frac{\pi d}{8\epsilon L} \right]^{1/2} \right] ,$$
 (23b)

where  $f_A = \dot{\gamma} n^2 d^2 / 2L^2$  and  $\epsilon$  is the area fraction of the particles. Equation (23) gives the standard error which is implicit in the use of gradient theory for regular flows. For fixed  $\epsilon$ , the error decreases with system size (L/d); however, it can be quite significant for small  $\epsilon$  (e.g., error  $\approx 20\%$  for  $\epsilon = 0.01$ , L = 10 cm, and  $d = 100 \, \mu$ m).

Two-dimensional chaotic flows, in general, exhibit islands and cannot be ergodic since the islands form an invariant set of finite measure.<sup>10</sup> (We note that the net flux across any cantorus must be zero by area conservation<sup>11</sup> so that cantori would not contribute to nonergodicity of the flow.) Trajectories in the chaotic region of the flow have positive Liapunov exponents, and the flow in these regions is mixing. Consequently,  $\psi_t$  is ergodic in the chaotic regions with the islands excluded, and  $\langle f \rangle_{\infty} = f_A$  independent of the number density. If the area occupied by the islands is sufficiently small,  $\langle f \rangle_{\infty}$ would approach  $f_A$  for chaotic flows even in the limit of very low number densities. Any deviation between the two would be essentially due to the trapping of some par-

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ticles within island chains, and the exclusion of the rest from the islands. Collisions may occur between a particle trapped within an island chain and those in the chaotic region; however, collisions between two particles trapped in different island chains would generally not be possible. Analytical treatment of the problem is extremely difficult since, in general, there is a large number of islands of different periodicities. These effects are screened out with increasing number density, and at sufficiently high number densities the complex structure would play no role in determining the collision rate.

#### COMPUTATIONS

A number of particles (6-3000) were placed on a grid in the chaotic region. The particles were mapped for several cycles (100-5000) to distribute them uniformly over the chaotic region. The average collision rate was then computed by counting the collisions in each cycle for a number of cycles. A collision was said to occur if the particle centers approached each other closer than the collision diameter (d). Only pairwise collisions were counted.

Computations were carried out to study (i) the parametric dependence of the collision frequency for very dilute suspensions (n = 6-12), and (ii) the crossover from dilute to nondilute suspensions keeping the system parameters fixed. Computations in (ii) were carried out at  $\mu = 0.4$ , which is close to  $\mu_c$ , and  $\mu = 1.0$ .

## **RESULTS AND DISCUSSION**

In all the cases that we studied the average collision rate approached an asymptotic value which was nearly independent of the interval of averaging. The rate of approach to an asymptotic value for the dilute suspensions was studied by considering the mean-square deviation of the collision frequency averaged over N cycles  $(\langle f \rangle_N)$  from its asymptotic value  $(\langle f \rangle_\infty)$  as

$$\sigma_N^2 = \overline{(\langle f \rangle_N - \langle f \rangle_\infty)^2} , \qquad (24)$$

where the overbar represents the average over several runs of duration N. The asymptotic collision frequency for the dilute suspensions was taken to be the average computed over 30 000 cycles since the fluctuations were very small beyond this value. The decay of the mean-square deviation was found to be

$$\frac{\sigma_N^2}{\langle f \rangle_{\infty}^2} = C_1 / N \tag{25}$$

(Fig. 3), and a least-square-fit line indicated that  $C_1 \propto n^{-2}$ . The dependence on  $\mu$  and d was found to be more complex and is shown in Fig. 4. The rate of convergence decreases with increasing flow strength and decreasing particle diameter.

The above results indicate that the interval of averaging required to approach  $\langle f \rangle_{\infty}$  within a fixed accuracy decreases with increasing number of particles (n), as expected. When the number density is high enough so that convergence of the asymptotic value with a given accura-



FIG. 3. Graph of deviation of  $\langle f \rangle_N (\sigma_N^2 / \langle f \rangle_\infty^2)$  vs 1/N for  $\mu = 1.0$ , d = 0.6, for different number of particles n = 6 ( $\Box$ ), n = 8 ( $\triangle$ ), n = 10 ( $\diamondsuit$ ), and n = 12 (+).



FIG. 4. Dependence of rate of convergence  $(C_1)$  on system parameters  $(\mu, d)$ : (a)  $C_1$  vs  $\mu$  for n = 10, d = 0.6; and (b)  $C_1$  vs d for  $\mu = 1.0$ , n = 10.

cy occurs in one cycle of the flow, the flow is essentially a regular flow (i.e., a shear flow due to a point vortex). For  $\mu = 1.0$  and d = 0.06, the number of particles required for  $\sigma_1/\langle f \rangle_{\infty} = 0.01$  (in one cycle) is  $n_c = 1250$  using Eq. (25) and  $C_1 \propto n^{-2}$ . While a considerable extrapolation is required in the above calculation,  $n_c$  gives an estimate of the number density at which the crossover to the nondilute regime occurs.

## Parametric dependence

The dependence of  $\langle f \rangle_{\infty}$  on *d* and *n* for the dilute solutions was found to be qualitatively similar to that of nondilute suspensions  $(\langle f \rangle_{\infty} \propto n^2 d^2)$  when  $\mu$  was sufficiently large.<sup>12</sup> The behavior for  $\mu = 0.4$  was somewhat different with

$$\langle f \rangle_{\infty} = n^{2.23} d^2 . \tag{26}$$

In fact, in all the cases there is a slow variation with n in addition to the  $n^2$  dependence.

The normalized collision frequency defined as

$$\langle f \rangle_{\infty}^{*} = \frac{\langle f \rangle_{\infty} / A}{\rho^2 d^2} \tag{27}$$

is shown in Fig. 5 and was found to increase with increasing flow strength. The increase in area with  $\mu$  $(A \propto \mu^{0.5-0.8})$  results in the particles exploring regions of relatively lower shear rate. At the same time the absolute shear rate increases at a rate proportional to  $\mu$ . Thus there is an effective increase in the average shear rate, leading to an increased rate of collisions. The rate of unmixing thus increases with flow strength in contrast to the average stretching rate (Liapunov exponent), which shows a maximum value at  $\mu \approx 1.^8$ 

## Crossover from the dilute to nondilute regime

The collision frequencies were calculated over a wide range of number densities to study the approach of the



FIG. 5. Graph of the normalized collision frequency  $\langle f \rangle_{\infty}^{*} = (\langle f \rangle_{\infty} / A \rho^{2} d^{2})$  as a function of the flow strength ( $\mu$ ) for n = 6 ( $\Diamond$ ), n = 8 ( $\triangle$ ), n = 10 ( $\Box$ ), and n = 12 ( $\times$ ).

asymptotic collision rate to the area averaged value, and the ratio  $f_{\infty}/f_A$  is shown in Fig. 6. Following Muzzio and Ottino,<sup>6</sup>  $f_A$  was found by evaluating the area integral [Eq. (16)] over two circles centered on the vortices, with the area equal to the area of the chaotic region. The area averaged frequency is then

$$f_A = c\rho^2 d^2 \int_L \dot{\gamma} \, dA \quad , \tag{28}$$

where c is fitted by matching  $f_A$  to  $\langle f \rangle_{\infty}$  at high number densities and L represents two circles. In our computations we obtained c = 0.82 for both  $\mu = 0.4$  and 1.0. Muzzio and Ottino<sup>6</sup> obtained  $c \approx 1.0$ .

The results show a significant deviation of  $\langle f \rangle_{\infty} / f_A$ from unity at very low number densities: for  $\mu = 0.4$  the maximum deviation calculated was about 25% and for  $\mu = 1.0$ , a maximum of about 12%. As discussed earlier, the higher value of deviation cannot be due to the presence of cantori, but is due to a greater fraction of the area covered by islands at lower flow strengths. In both the cases the time-averaged frequency is lower than the area averaged value at low number densities. This is expected since the trapping of particles in islands would reduce the effective concentration in the chaotic region and thus the collision frequency.

At both flow strengths,  $\langle f \rangle_{\infty}$  approaches  $f_A$  rapidly with increasing  $\rho$ , and the deviation is less than 5% for  $\rho \approx 5.2$ . This implies that in the case of chaotic flows, the gradient theory is reasonably accurate even at later stages of the aggregation process when the number density is low. In contrast, the deviations would be much greater in the case of a regular flow at the same number density. The convergence does not seem to be uniform with further increase in number density; however, at  $n_c \approx 1000$ the deviation of  $\langle f \rangle_{\infty}$  from  $f_A$  is less than 2% (i.e., the error of computation). For this number density  $\langle f \rangle_{\infty}$  is reached within one time period of averaging. This value is in reasonable agreement with the predictions based on the results of the preceding section.



FIG. 6. Ratio of the asymptotic collision frequency  $(\langle f \rangle_{\infty})$  to the area averaged value  $(f_A)$  vs the number density  $(\rho)$  for  $\mu = 1.0$  ( $\Box$ ) and  $\mu = 0.4$  ( $\triangle$ ); d = 0.06.

## CONCLUSIONS

We have computed to a high accuracy the collision rates in very dilute suspensions subject to the blinking vortex flow. Our computations show that the long time average of the collision frequency achieves an asymptotic value which is nearly independent of the initial positions of the particles, and at high flow strengths the behavior for dilute suspensions is similar to the nondilute case. In addition, with increasing number density there is a rapid convergence of  $\langle f \rangle_{\infty}$  to the prediction of the gradient theory. The latter is due to the mixing property of the flow over a large fraction of the chaotic region, and would be expected to hold for chaotic flows in general. Thus, as suggested by Muzzio and Ottino,<sup>6</sup> the gradient theory is more accurate for chaotic flows as compared to regular flows for all number densities.

The results presented here show a complex dependence of the collision frequency on the flow at low number densities. The behavior is a reflection of the structure underlying the flow. In the limit of two particles of small diameter, the ratio  $\langle f \rangle_{\infty} / f_A$  can be considered to be a global measure which is indicative of mixing property of the flow. For flows which are mixing  $\langle f \rangle_{\infty} / f_A = 1$ , and for flows which are near mixing, this ratio would be less than 1. The diameter of the particles would specify the length scale to be probed. The measure is complementary to the Liapunov exponent, which is a local measure of stretching of material lines in the flow. A detailed characterization of single-pair collision rates for model flows is planned.

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