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Fluctuations for annihilations of Brownian spheres

Mathew D. Penrose*

Department of Statistics and Applied Probability, University of California, Santa Barbara, CA 93106-3110, USA

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Abstract

As a model for a diffusion-limited chemical reaction, we consider a large number N of spheres, of small radius r_N, which perform independent Brownian motions in Euclidean space and annihilate one another on contact. We consider the point process of annihilations, and show that according to the limiting behavior of r_N, this point process may converge weakly, either to a Poisson process or (after re-normalization) to white noise.

Key words: Chemistry, Reaction, Diffusion, Brownian motion, Central limit theorem, White noise, Poisson process, U-statistics.

1. Introduction

Consider a reaction–diffusion system, consisting of N Euclidean balls of diameter r_N in $\mathbb{R}^d$, $d \geq 2$, whose centers perform independent Brownian motions, with independent identically distributed (i.i.d.) initial positions with a bounded density function $u(x)$, $x \in \mathbb{R}^d$. Suppose that whenever two balls collide, they disappear. Also, let any particle initially placed within a distance r_N of any other, be removed from the system at once.

This system is a model for the chemical reaction A + A → P, where P is an inert product. Imagine each ball to be a molecule of substance A, which executes Brownian motion in a suspension fluid until it touches another molecule of A. As soon as this happens, the two A molecules which touch are replaced by a molecule of P, which undergoes no further reaction. The reaction is diffusion-limited; the reaction of molecules is instantaneous, while their motion is not. For surveys of related models, see Clifford et al. (1987) and Kotelenez (1986, 1988).

We shall consider the limiting behaviour of the system as $N \to \infty$, when (r_N) is a given sequence converging to zero. Define the function S_d on $(0, \infty)$ by

$$S_d(x) = \log(1/x) \quad (d = 2),$$

* Correspondence address: Department of Mathematical Sciences, University of Durham Science Site, South Road, Durham DH1 3LE, UK.

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$$S_d(x) = x^{2-d} \quad (d \geq 3),$$

and define the sequence (s_N) by

$$s_N = (S_d(r_N/\sqrt{2}))^{-1}. \tag{1.1}$$

The case $Ns_N = \text{const.}$ was studied by Sznitman (1987), who proved a propagation of chaos result for the density of particles. This case is of interest because the number of collisions before time 1 grows in proportion to N as $N \to \infty$.

In this paper, we study the $N \to \infty$ limiting behaviour of a point process η_N on $\mathbb{R}_+ \times \mathbb{R}^d$, obtained by recording the time and place of each 'reaction' (annihilation), that is by recording the time and place of the creation of each inert P molecule. This approach differs from that of papers on related models, such as Sznitman (1987), Lang and Xanh (1980), Dittrich (1988), Nappo and Orlandi (1988), Nappo et al. (1989) and Kotelenez (1991), who considered instead the evolving system of surviving particles.

In contrast with the case $Ns_N = \text{const.}$, we here consider cases when $Ns_N \to 0$ but $N^2 s_N$ is bounded away from 0 as $N \to \infty$. In these cases, the number of collisions before time 1 becomes much smaller than N; our point process approach allows the study of the annihilations even when their number is swamped by the number of surviving particles. We shall obtain Poisson limits when $N^2 s_N \to \text{const.}$, and Gaussian limits (after re-normalization) when $N^2 s_N \to \infty$.

2. Definitions

Let $d \geq 2$ be an integer. Let $(u(x), x \in \mathbb{R}^d)$ be a bounded probability density function. On a probability space $(\Omega_N, \mathscr{F}_N, P_N)$, let $(X_i(t), t \geq 0)$, $1 \leq i \leq N$, be independent standard Wiener processes in $\mathbb{R}^d$, with initial distribution $P[X_i(0) \in dx] = u(x)\,dx$. (In this paper, 'Brownian motion' denotes a physical process, and the mathematical object usually given that name is denoted a 'Wiener process'.) Note that $X_i(t)$ runs for all $t > 0$ even after the annihilation of the corresponding particle. For distinct $i, j \in \{1, 2, \ldots, N\}$, set

$$Y_{ij}(t) = (X_i(t) - X_j(t))/\sqrt{2},$$

$$Y_{ij}(t) = (X_i(t) + X_j(t))/\sqrt{2}.$$

Let (r_N) be a sequence of strictly positive numbers. Let the sequence (s_N) be defined by (1.1). Set

$$T_{ij} = \inf\{t \geq 0: |Y_{ij}(t)| \leq r_N/\sqrt{2}\},$$

where $|\cdot|$ denotes the Euclidean modulus. Note that T_{ij} depends on N. Then $\{T_{ij}: 1 \leq i < j \leq N, T_{ij} > 0\}$ are distinct, since for $i' \neq i, j$, the distribution of $X_{i'}(T_{ij})$ has a density, and so for $j' \neq i'$, by sample path continuity

$$P[T_{i'j'} = T_{ij} > 0] \leq P[|Y_{i'j'}(T_{ij})|r_N/\sqrt{2}] = 0.$$

Following Section 2 of Sznitman (1987), let particles i and j be 'annihilated' at time T_{ij}, provided neither particle was annihilated at an earlier time. Let T^k be the kth time at which an annihilation takes place. Then $0 \leq T^1 < T^2 < \cdots < T^L$, where L is the (random, finite) total number of such times. Also, if $k \leq L$ and $T^k > 0$, then the annihilation at time T^k involves exactly two particles; denote their (random) indices $i(k)$ and $j(k)$, with $i(k) < j(k)$ (so $T_{i(k),\, j(k)} = T^k$). Set

$$Z^k = \tilde{Y}_{i(k),\, j(k)}(T^k).$$

Then $Z^k/\sqrt{2}$ is the place at which the collision at time T^k occurs.

Let η_N be the point process on $\mathbb{R}_+ \times \mathbb{R}^d$ with points at (T^k, Z^k), $(k \leq L,\ T^k > 0)$. That is, for any test function $f: \mathbb{R}_+ \times \mathbb{R}^d \to \mathbb{R}$, set

$$\eta_N(f) = \sum_{k=1}^{L} f(T^k, Z^k) I_{\{T^k > 0\}}, \tag{2.1}$$

where $I_{\{\ \}}$ denotes the indicator function, and for any $R \subset \mathbb{R}_+ \times \mathbb{R}^d$, define $\eta_N(R)$ by

$$\eta_N(R) := \eta_N(I_R),$$

where $:=$ denotes definition and I_R denotes the characteristic function of R.

Let $v(y, \tilde{y})$ be the version of the joint density of $(Y_{12}(0), \tilde{Y}_{12}(0))$, given by

$$v(y, \tilde{y}) := u((\tilde{y} + y)/\sqrt{2})\, u((\tilde{y} - y)/\sqrt{2}).$$

Let $p_t(x)$ $(t > 0,\ x \in \mathbb{R}^d)$ denote the Brownian transition density; that is, $p_t(x) := (2\pi t)^{-d/2} \exp(-|x|^2/2t)$.

Set $\pi_d := \pi^{d/2}\,\Gamma((d/2) + 1)^{-1}$, the volume of the unit ball in $\mathbb{R}^d$. Define C_d by

$$C_d := \pi \qquad (d = 2),$$

$$C_d := \left(\frac{d}{2} - 1\right) d\pi_d \quad (d \geq 3).$$

Define the function λ on $\mathbb{R}_+ \times \mathbb{R}^d$ by

$$\lambda(t, x) = C_d \int_{\mathbb{R}^d} \int_{\mathbb{R}^d} p_t(x - \tilde{y})\, p_t(y)\, v(y, \tilde{y})\, \mathrm{d}y\, \mathrm{d}\tilde{y}, \quad (t, x) \in \mathbb{R}_+ \times \mathbb{R}^d. \tag{2.2}$$

Note that $\lambda(t, x)$ is a constant times the continuous version of the joint density of $(Y_{12}(t), \tilde{Y}_{12}(t))$ at $(0, x)$.

Let $\xrightarrow{\mathrm{d}}$ denote convergence in law.

3. Statement of results

3.1. Poisson limit theorems

First, we consider the case when $N^2 s_N$ converges to a finite non-zero limit. Let $\mathscr{R}_{d+1}$ denote the ring of all finite unions of sets in $\mathbb{R}_+ \times \mathbb{R}^d$ of the form $J \times A$, where $J \subset \mathbb{R}_+$ is a bounded interval and A is a Borel subset of $\mathbb{R}^d$.

Theorem 1. *Suppose $N^2 s_N \to 2\gamma \in (0, \infty)$ as $N \to \infty$. Then for all $R \in \mathscr{R}_{d+1}$, $\eta_N(R)$ converges in law to a Poisson random variable with mean $\gamma \int_R \lambda(t, x)\,\mathrm{d}t\,\mathrm{d}x$.*

Let the space of point measures on $\mathbb{R}_+ \times \mathbb{R}^d$ have the vague topology; that is, $a_n \to a \Leftrightarrow a_n f \to af$, $f \in C_0(\mathbb{R}_+ \times \mathbb{R}^d)$. By Kallenberg (1973, Theorem 2.3), we have convergence in law of η_N to a Poisson process:

Corollary 1. *Suppose $N^2 s_N \to 2\gamma$ as $N \to \infty$, with $0 < \gamma < \infty$. Then the random point measure η_N converges in law to a Poisson process with mean measure $\gamma\lambda(t, x)\,\mathrm{d}x\,\mathrm{d}t$.*

3.2. Gaussian limit theorems

If $N^2 s_N \to \infty$, then we must re-normalize η_N to obtain a limit law. Define the signed measure ζ_N on test functions f on $\mathbb{R}_+ \times \mathbb{R}^d$ by

$$\zeta_N(f) = (N^2 s_N/2)^{-1/2}(\eta_N(f) - E_N \eta_N(f)), \tag{3.1}$$

(recall, E_N is the expectation corresponding to P_N). For any set R in $\mathscr{R}_{d+1}$, define $\zeta_N(R)$ by identifying R with its characteristic function.

The limit we shall obtain is white noise, denoted W, on $\mathbb{R}_+ \times \mathbb{R}^{d+1}$, with intensity measure $\lambda(t, x)\,\mathrm{d}x\,\mathrm{d}t$. This is defined to be a set-indexed, centered Gaussian process $(W(R, \omega), R \in \mathscr{R}_{d+1}, \omega \in \Omega)$ on some probability space $(\Omega, \mathscr{F}, P)$, with

$$\mathrm{Cov}(W(R), W(R')) = \int_{R \cap R'} \lambda(t, x)\,\mathrm{d}t\,\mathrm{d}x.$$

Roughly, W is the Gaussian equivalent of a Poisson process. Viewing ζ_N as a (generalized) process indexed by sets in $\mathscr{R}_{d+1}$ (we were unable to obtain results on any larger class of sets), we have the following theorem.

Theorem 2. *Suppose $N^2 s_N \to \infty$ and for some $\varepsilon > 0$, $N^{1+\varepsilon} s_N \to 0$ as $N \to \infty$. Then the finite-dimensional distributions of the process $(\zeta_N(R), R \in \mathscr{R}_{d+1})$ converge to those of*

$(W(R), R \in \mathscr{R}_{d+1})$. *That is, for* $R_1, \ldots, R_n \in \mathscr{R}_{d+1}$,

$$(\zeta_N(R_1), \ldots, (\zeta_N(R_n)) \xrightarrow{\mathrm{d}} (W(R_1), \ldots, W(R_n)) \quad \text{as } N \to \infty .$$

Another interpretation is to work in the space $\mathscr{S}'$ of tempered distributions on $\mathbb{R}^{d+1}$; that is, the topological dual of the space $\mathscr{S}$ of rapidly decreasing functions on $\mathbb{R}^{d+1}$, endowed with the strong topology; see for example Walsh (1986) for details. In this setting, view η_N and ζ_N, given by (2.1) and (3.1), as random elements of $\mathscr{S}'$. As for white noise, let $(W(f), f \in \mathscr{S})$ be a centered Gaussian generalized process with

$$\mathrm{Cov}(Wf, Wg) = \int_{\mathbb{R}^{d+1}} f(t,x)\, g(t,x)\, \lambda(t,x)\, \mathrm{d}x\, \mathrm{d}t.$$

We can and do take a version of $(W(f,\omega), f \in \mathscr{S}, \omega \in \Omega)$ on some probability space $(\Omega, \mathscr{F}, P)$ such that $W(\cdot, \omega) \in \mathscr{S}'$ for P-almost every $\omega \in \Omega$. See Walsh (1986, Theorem 4.1).

Here we study only weak convergence on bounded time intervals. For each $\tau > 0$ define the random distributions η_N^τ, ζ_N^τ and W^τ to be the restrictions of η_N, φ_N and W, respectively, to $(0, \tau] \times \mathbb{R}^d$. That is, set

$$\eta_N^\tau(f) := \eta_N(f(\cdot)\, I_{(0,\tau] \times \mathbb{R}^d}(\cdot)),$$

and define ζ_N^τ similarly; set W^τ to be white noise with intensity measure $\lambda(t,x) \times I_{(0,\tau]}(t)\, \mathrm{d}x\, \mathrm{d}t$. In this setting, the result is given by the following theorem.

Theorem 3. *Suppose* $N^2 s_N \to \infty$ *and for some* $\varepsilon > 0$, $N^{1+\varepsilon} s_N \to 0$ *as* $N \to \infty$. *Then for all* $\tau > 0$, $\zeta_N^\tau \xrightarrow{\mathrm{d}} W^\tau$ *in* $\mathscr{S}'$ *as* $N \to \infty$.

Theorems 2 and 3 are not entirely satisfactory, since in the expression (3.1) for ζ_N, the constant to be subtracted from η_N is not explicitly stated in terms of the initial density function $u(\cdot)$. When $d = 3$ and $N^{4/3} s_N \to 0$, we can be more explicit. For $\tau > 0$, define the random element ξ_N^τ of $\mathscr{S}'$ by

$$\xi_N^\tau(f) = (N^2 s_N/2)^{-1/2} \left\{ \eta_N^\tau(f) - \binom{N}{2} s_N \int_0^\tau \int_{\mathbb{R}^d} f(t,x)\, \lambda(t,x)\, \mathrm{d}x\, \mathrm{d}t \right\} \tag{3.2}$$

$$= \zeta_N^\tau(f) + (N^2 s_N/2)^{-1/2} \left\{ E_N\, \eta_N^\tau(f) - \binom{N}{2} s_N \int_0^\tau \int_{\mathbb{R}^d} f(t,x)\, \lambda(t,x)\, \mathrm{d}x\, \mathrm{d}t \right\}. \tag{3.3}$$

Theorem 4. *Suppose $d = 3$, $N^2 s_N \to \infty$ and $N^{4/3} s_N \to 0$. Then for all $\tau > 0$, $\xi_N^\tau \xrightarrow{\mathrm{d}} W^\tau$ in $\mathscr{S}'$ as $N \to \infty$.*

It is harder to obtain an analogous improvement to Theorem 2 (characteristic functions are harder to work with than functions in $\mathscr{S}$). Here, we content ourselves with considering only sets $R \subset \mathbb{R}^{d+1}$ of the form $R = J \times \mathbb{R}^d$, where J is a finite union of intervals. This amounts to looking only at the finite-dimensional distributions of a stochastic process with time parameter $t \in [0, \infty)$, obtained by counting the total number of annihilations before time t. The re-normalized process, which we denote $(\psi_N(t), t \geq 0)$ is given by

$$\psi_N(t) = \xi_N^t(1)$$

$$= (N^2 s_N/2)^{-1/2} \left\{ \eta_N((0,t] \times \mathbb{R}^d) - \binom{N}{2} s_N \int_0^t \int_{\mathbb{R}^d} \lambda(s,x)\,\mathrm{d}x\,\mathrm{d}s \right\} \tag{3.4}$$

$$= \zeta_N((0,t] \times \mathbb{R}^d) + (N^2 s_N/2)^{-1/2} \left\{ E\eta_N((0,t] \times \mathbb{R}^d) - \binom{N}{2} s_N \int_0^t \int_{\mathbb{R}^d} \lambda(s,x)\,\mathrm{d}x\,\mathrm{d}s \right\}. \tag{3.5}$$

Observe that in (3.4), the constant subtracted from η_N to get ψ_N is explicitly stated in terms of $u(\cdot)$, as in the case of the expression (3.2) for ξ_N^τ.

The limit process in this setting is white noise W_1 on $[0, \infty)$, with intensity $\lambda_1(t)\,\mathrm{d}t$ given by

$$\lambda_1(t) := \int_{\mathbb{R}^d} \lambda(t,x)\,\mathrm{d}x$$

$$= C_d \int_{\mathbb{R}^d} \int_{\mathbb{R}^d} p_t(y) v(y,\tilde{y})\,\mathrm{d}y\,\mathrm{d}\tilde{y}, \quad t > 0.$$

But $W_1(t) := W_1([0,t])$ is just a time-changed Wiener process in $\mathbb{R}$, starting at 0. So a natural statement of the result is as follows.

Theorem 5. *If $d = 3$, $N^2 s_N \to \infty$ and $N^{4/3} s_N \to 0$, then the finite-dimensional distributions of the process $(\psi_N(t), t \geq 0)$ converge to those of a process $(W_1(t), t \geq 0)$ given by*

$$W_1(t) = B\Big(\int_0^t \lambda_1(s)\,\mathrm{d}s\Big),$$

where $(B(t), t \geq 0)$ is a one-dimensional Wiener process starting at 0.

Theorems 4 and 5 can be extended to some higher-dimensional cases. We omit these results for the sake of brevity. See below for a remark on a possible extension of theorems 4 and 5 beyond the case where $N^{4/3} s_N \to 0$.

The following are the main ideas in the proofs to follow. First, we approximate to η_N^τ by a point process ϕ_N^τ obtained by simply recording each $(T_{ij}, \tilde{Y}_{ij}(T_{ij}))$, ignoring the question of whether particle i or j has already been annihilated by time T_{ij}. That is, for $\tau > 0$ and all test functions f on $\mathbb{R}_+ \times \mathbb{R}^d$, set

$$\phi_N^\tau(f) := \sum_{1 \le i < j \le N} \sum f(T_{ij}, \tilde{Y}_{ij}(T_{ij})) I_{\{0 < T_{ij} \le \tau\}}. \tag{3.6}$$

But $\phi_N^\tau(f)$ has the form of a U-statistic; that is, a sum over all distinct pairs taken from N i.i.d. E-valued random variables, of a given function on $E \times E$, where E is the measurable space $C([0, \infty), \mathbb{R}^d)$. We use limit theorems on U-statistics found in Jammalamadaka and Janson (1986) or elsewhere. Those results are stated in the case when E is Euclidean space, but the proofs carry over to the case where E is an arbitrary measurable space.

To apply these results, we need to know about the limit behaviour of the law of T_{12} as r_N becomes small. Such results are to be found in Le Gall (1986).

The mean number of particles which would collide before time τ with two distinct others, if the annihilation reaction were 'switched off', is of the order of $N^3 s_N^2$, since the probability that a specified particle collides with both of two other specified particles is $O(s_N^2)$. The renormalization in (3.1) involves dividing by $(N^2 s_N)^{1/2}$. Therefore, when $N^{4/3} s_N \to 0$, $\phi_N^\tau(f)$ is a good approximation to $\eta_N^\tau(f)$, since in this case $(N^3 s_N^2)/(N^2 s_N)^{1/2}$ approaches 0. Otherwise we must estimate the variance of the error caused by approximating to η_N^τ by ϕ_N^τ. We do this in Section 8, studying the combinatorics of a series of collisions by a graph-theoretic method.

When $d = 3$, it may be possible to use the graph-theoretic method to obtain an approximation for $E_N \eta_N^\tau(f)$ in terms of the initial density u, and thus to extend Theorems 4 and 5 beyond the case $N^{4/3} s_N \to 0$. However, the approximation to $E_N \eta_N^\tau(f)$ will be much more complicated in the general case than it is in Eq. (3.3) for the case $N^{4/3} s_N \to 0$; it will be a sum over graphs.

The use of U-statistics should be applicable in some of the related models discussed in the papers referred to in Section 1. The method can also be applied to a model with two types of particles, A and B, for the reaction A + B → P, with P inert. See Penrose (1992).

In the proofs to follow, c denotes a finite positive constant, and may change from line to line.

4. Preliminary results

Lemma 1. *Suppose $\tau > 0$ and $f \in L^\infty(\mathbb{R}_+ \times \mathbb{R}^d)$. Then*

(i) *if $N^{3/2} s_N \to 0$ then*

$$P_N[\phi_N^\tau(f) = \eta_N^\tau(f)] \to 1 \quad as\ N \to \infty,$$

(ii) *if* $N^{4/3}s_N \to 0$ *then*

$$(N^2 s_N)^{-1/2} E_N[|\phi_N^\tau(f) - \eta_N^\tau(f)|] \to 0 \quad \text{as } N \to \infty .$$

Proof. Suppose $1 \le i < j \le N$. Only when $T_{ik} \le T_{ij} \le \tau$ or $T_{kj} \le T_{ij} \le \tau$ for some $k \ne i,j$, does the contribution of pair (i,j) to $\eta_N^\tau(f)$ differ from its contribution to $\phi_N^\tau(f)$. That is,

$$|\phi_N^\tau(f) - \eta_N^\tau(f)| \le \|f\|_\infty \sum\sum\sum I_{\{T_{ik} \le T_{ij} \le \tau\}}$$

where the sum runs through all distinct i,j and k in $\{1, 2, \ldots, N\}$. The result follows from the fact that

$$P_N[T_{12} \le T_{13} \le \tau] = O(s_N^2) \quad \text{as } N \to \infty . \tag{4.1}$$

See the proof of (6.1) of Penrose (1991), or Proposition 2. □

The next result is based on the limiting expression for the probability that a Wiener process in $\mathbb{R}^d$ hits a small ball, found in Le Gall (1986).

Lemma 2. *Suppose* $h \in L^\infty(\mathbb{R}_+ \times \mathbb{R}^d)$, *and for some* $\tau > 0$, *for all* $\tilde{y} \in \mathbb{R}^d$, $h(t, \tilde{y}) = 0$ *for* $t > \tau$ *and* $h(\cdot, \tilde{y})$ *is piecewise continuous (the intervals of continuity may depend on* $\tilde{y}$*). Then*

$$\lim_{N \to \infty} s_N^{-1} E_N h(T_{12}, \tilde{Y}_{12}(0)) = C_d \int_{\mathbb{R}^d} \int_{\mathbb{R}^d} \int_0^\infty h(t, \tilde{y}) p_t(y) v(y, \tilde{y}) \, dt \, dy \, d\tilde{y} \tag{4.2}$$

and the limit in (4.2) *is finite.*

Proof. By definition, we have

$$E_N h(T_{12}, \tilde{Y}_{12}(0)) = \int_{\mathbb{R}^d} \int_{\mathbb{R}^d} E^y[h(T_N, \tilde{y})] \, v(y, \tilde{y}) \, dy \, d\tilde{y} \tag{4.3}$$

where E^y is expectation (and P^y is probability) with respect to a Brownian motion $(Y(t), t \ge 0)$ starting at y, and

$$T_N = \inf\{t : |Y(t)| \le r_N/\sqrt{2}\}.$$

Fix $\tilde{y}$ and y for the moment, with $y \ne 0$. Suppose $h(\cdot, \tilde{y})$ is the characteristic function of an interval. Then by Corollaire 1-2 of Le Gall (1986),

$$s_N^{-1} E^y h(T_N, \tilde{y}) \to \int_0^\infty C_d h(t, \tilde{y}) p_t(y) \, dt \quad \text{as } N \to \infty . \tag{4.4}$$

Also, the limit (4.4) still holds if $h(\cdot, \tilde{y})$ is a step function, by linearity. Finally, if $h(\cdot, \tilde{y})$ is piecewise continuous, it is Riemann integrable; approximating to h from above and below by step functions, we may deduce that (4.4) still holds, using the fact that $p_t(y)$ is bounded on $\{0 < t \leq \tau\}$.

We can also now deduce (4.2), provided we can find a suitable function to dominate the function $s_N^{-1} E^y h(T_N, \tilde{y})$. By the majorization of Le Gall (1986), Lemme 2-1, and routine use of Brownian scaling to account for the possibility that $\tau > 1$, we have for some c and N_0 and all $N \geq N_0, y$ and $\tilde{y}$ in $\mathbb{R}^d$:

$$s_N^{-1} | E^y h(T_N, \tilde{y})| \leq \|h\|_\infty s_N^{-1} P^y[T_N \leq \tau] \leq c f_d(\tau^{-1/2} |y|) \tag{4.5}$$

where

$$f_d(x) := (S_d(x)_+ + 1) \exp(-x^2/16) \tag{4.6}$$

and $a_+ := \max(a, 0)$ is the positive part of a. Finally, by Hölder's inequality we have

$$\int_{\mathbb{R}^d} v(y, \tilde{y}) \, \mathrm{d}\tilde{y} \leq \sqrt{2} \|u\|_2 < \infty, \tag{4.7}$$

(since the density u was assumed bounded), and so

$$\int_{\mathbb{R}^d} \int_{\mathbb{R}^d} f_d(\tau^{-1/2}|y|) v(y, \tilde{y}) \, \mathrm{d}\tilde{y} \, \mathrm{d}y \leq \sqrt{2} \|u\|_2 \int_{\mathbb{R}^d} f_d(\tau^{-1/2}|y|) \, \mathrm{d}y < \infty.$$

The result (4.2) follows by (4.5) and Dominated Convergence. Also, by (4.7), the assumptions on h and the fact that $\|p_t\|_1 = 1$, the limit in (4.2) is finite. □

5. Proof of Theorem 1.

Let $R \in \mathscr{R}_{d+1}$. Take $\tau > 0$ so that $R \subset [0, \tau] \times \mathbb{R}^d$. By Lemma 1, it is enough to prove that

$$\phi_N^\tau(R) \xrightarrow{\mathrm{d}} \text{Poisson} \left(\gamma \int_{\mathbb{R}} \lambda(t, x) \, \mathrm{d}t \, \mathrm{d}x \right). \tag{5.1}$$

Define the function f on $\mathbb{R}_+ \times \mathbb{R}^d$ to be the characteristic function of R (so $f^2 \equiv f$). We have

$$\phi_N^\tau(R) = \sum_{1 \leq i < j \leq N} \sum U_{ij}$$

where

$$U_{ij} := f(T_{ij}, \tilde{Y}_{ij}(T_{ij})) I_{\{0 < T_{ij} \leq \tau\}}. \tag{5.2}$$

Since $Y_{12}(\cdot) - Y_{12}(0)$ and $\tilde{Y}_{12}(\cdot) - \tilde{Y}_{12}(0)$ are independent Wiener processes starting at 0, we have

$$E_N[U_{12}^2] = E_N[E_N(U_{12}^2 | T_{12}, \tilde{Y}_{12}(0))] = E_N h(T_{12}, \tilde{Y}_{12}(0)),$$

where we set

$$h(t, \tilde{y}) = I_{(0,\tau]}(t) \int_{\mathbb{R}^d} f^2(t, x) p_t(x - \tilde{y}) \, dx, \quad (t, \tilde{y}) \in [0, \infty] \times \mathbb{R}^d.$$

Since $f \in L^\infty(\mathbb{R}_+ \times \mathbb{R}^d)$, and $f(\cdot, x)$ is piecewise continuous for each x, the function h is piecewise continuous in t, bounded, and of bounded support. By Lemma 2,

$$\begin{aligned}\lim_{N \to \infty} s_N^{-1} E_N[U_{12}^2] &= C_d \int_{\mathbb{R}^d} \int_{\mathbb{R}^d} \int_0^\infty h(t, \tilde{y}) p_t(y) v(y, \tilde{y}) \, dt \, dy \, d\tilde{y} \\ &= \int_{\mathbb{R}^d} \int_0^\tau f^2(t, x) \lambda(t, x) \, dt \, dx, \end{aligned} \tag{5.3}$$

by the definition (2.2) of λ. By the assumption $N^2 s_N \to 2\gamma$ and the definition of f, we have

$$\lim_{N \to \infty} (N^2/2) E_N[U_{12}^2] = \gamma \int_R \lambda(t, x) \, dt \, dx. \tag{5.4}$$

Also, by (4.1),

$$\lim_{N \to \infty} N^3 E_N[U_{12} U_{13}] = 0. \tag{5.5}$$

By (5.4), (5.5) and results on U-statistics (see Silverman and Brown (1978, Theorem A) or Jammalamadaka and Janson (1986, Theorem 3.1)), (5.1) holds. □

6. Proof of Gaussian limits when $N^{4/3} s_N \to 0$.

The following application of a theorem on U-statistics is the key to the Gaussian limit theorems. Recall that W^τ denotes white noise with intensity $\lambda(x, t) I_{(0,\tau]}(t) \, dx \, dt$. For $\tau > 0$ and $f \in L^\infty(\mathbb{R}_+ \times \mathbb{R}^d)$, define $\tilde{\zeta}_N^\tau(f)$ analogously to $\zeta_N(f)$, but with η_N^τ replaced by ϕ_N^τ:

$$\tilde{\zeta}_N^\tau(f) := (N^2 s_N/2)^{-1/2} [\phi_N^\tau(f) - E_N \phi_N^\tau(f)].$$

Proposition 1. *Suppose $N s_N \to 0$ and $N^2 s_N \to \infty$ as $N \to \infty$. Let $\tau > 0$. Then*

(i) *for any* $f \in L^\infty(\mathbb{R}_+ \times \mathbb{R}^d)$, *with* $f(\cdot, x)$ *piecewise continuous* (*the intervals of continuity may depend on* x), *all* $x \in \mathbb{R}^d$,

$$\tilde{\zeta}_N^\tau(f) \xrightarrow{\mathrm{d}} \mathit{Normal}\left(0, \int_0^\tau \int_{\mathbb{R}^d} f^2(t,x)\lambda(t,x)\,\mathrm{d}x\,\mathrm{d}t\right) \quad \text{as } N \to \infty. \tag{6.1}$$

(ii) *For any* $f_1, \ldots, f_n$ *in* $L^\infty(\mathbb{R}_+ \times \mathbb{R}^d)$, *with* $f_i(\cdot, x)$ *piecewise continuous, all* $x \in \mathbb{R}^d$, $1 \le i \le n$, *we have*

$$(\tilde{\zeta}_N^\tau(f_1), \tilde{\zeta}_N^\tau(f_2), \ldots, \tilde{\zeta}_N^\tau(f_n)) \xrightarrow{\mathrm{d}} (W^\tau f_1, W^\tau f_2, \ldots, W^\tau f_n) \quad \text{as } N \to \infty.$$

Proof. (i) Observe that

$$(N^2 s_N/2)^{-1/2}\phi_N^\tau(f) = \sum\sum_{1 \le i < j \le N} V_{ij}, \tag{6.2}$$

where $V_{ij} := (N^2 s_N/2)^{-1/2} U_{ij}$, with U_{ij} given by (5.2). By the estimate (4.1), we have for some $c > 0$,

$$N^3 E_N[V_{12} V_{13}] \le c N s_N \to 0 \quad \text{as } N \to \infty. \tag{6.3}$$

Also, by the proof of Theorem 1, (5.3) holds; that is,

$$\lim_{N \to \infty} (N^2/2) E_N[V_{12}^2] = \int_{\mathbb{R}^d} \int_0^\tau f^2(t,x)\lambda(t,x)\mathrm{d}t\,\mathrm{d}x. \tag{6.4}$$

Let F_N be the distribution function of V_{12}. Since $\|V_{12}\|_\infty \to 0$, the measure $(N^2/2)(t^2/(1+t^2))\mathrm{d}F_N(t)$ converges completely (in the sense of Loève (1963, page 178)) to a point mass at 0, of size given by the expression in (6.4).

Also, for some $c > 0$ (which may change from line to line),

$$\begin{aligned}(N^2/2)|E_N[V_{12}/(1+V_{12}^2)] - E_N V_{12}| &\le c N^2 E_N[|V_{12}|^3] \\ &\le c N^2 (N^2 s_N/2)^{-3/2} P_N[T_{12} \le \tau],\end{aligned}$$

which approaches 0 as $N \to \infty$, by Lemma 2 and the assumption that $N^2 s_N \to \infty$. Moreover, by (6.2),

$$|(N^2/2) E_N V_{12} - (N^2 s_N/2)^{-1/2} E_N \phi_N^\tau(f)| = (N/2)|E_N V_{12}|,$$

which also approaches 0 as $N \to \infty$. So

$$(N^2/2) E_N(V_{12}/(1+V_{12}^2)) - (N^2 s_N/2)^{-1/2} E_N \phi_N^\tau(f) \to 0 \quad \text{as } N \to \infty. \tag{6.5}$$

By (6.2)–(6.5) and Jammalamadaka and Janson (1986, Theorem 3.1), we obtain (6.1).

(ii) This result follows from part (i) by use of the Cramér–Wold device. See Billingsley (1968), Theorem 7.7. □

Proof of Theorems 2 and 3 when $N^{4/3}s_N \to 0$. Apply Proposition 1 to characteristic functions of sets in $\mathscr{R}_{d+1}$. By Lemma 1 and Billingsley (1968, page 28, problem 1), the case $N^{4/3}s_N \to 0$ of Theorem 2 is immediate.

As for Theorem 3 when $N^{4/3}s_N \to 0$, the same argument shows that for $f_1, \dots, f_n$ in $\mathscr{S}$, $(\zeta_N^\tau(f_1), \dots, \zeta_N^\tau(f_n))$ converges in law to $(W^\tau(f_1), \dots, W^\tau(f_n))$. The desired convergence in law in $\mathscr{S}'$ now follows from Mitoma's theorem. See for example Walsh (1986, Theorem 6.15), setting $X_N(\cdot)$ to be the constant $\mathscr{S}'$-valued process $X_N \equiv \zeta_N^\tau$. □

7. Proof of Theorems 4 and 5

In these results $d = 3$ so $s_N = r_N/\sqrt{2}$. The next two results are estimates on the rate of convergence in Lemma 2.

Lemma 3. *Suppose $d = 3$, $\tau > 0$. Then there are constants c and N_0, such that for all $t \le \tau$ and all bounded measurable functions $(a(\tilde{y}), \tilde{y} \in \mathbb{R}^3)$, for $N \ge N_0$,*

$$|s_N^{-1} E_N[a(\tilde{Y}_{12}(0)) I_{\{0 < T_{12} \le t\}}] - 2\pi \int_{\mathbb{R}^d} \int_{\mathbb{R}^d} \int_0^t a(\tilde{y}) p_s(y) v(y, \tilde{y}) \, \mathrm{d}s \, \mathrm{d}y \, \mathrm{d}\tilde{y}| \le c s_N \|a\|_\infty .$$

Proof. We may re-write (4.3) as follows:

$$E_N[a(\tilde{Y}_{12}(0)) I_{\{0 < T_{12} \le t\}}] = \int_{\mathbb{R}^d} \int_{|y| > s_N} a(\tilde{y}) P^y[T_N \le t] \, v(y, \tilde{y}) \, \mathrm{d}y \, \mathrm{d}\tilde{y} \tag{7.1}$$

where, under P^y, as before, T_N is the first time a Wiener process starting at y visits $\{|x| \le s_N\}$. When $d = 3$ there is an exact expression for $P^y[T_N \le t]$. See (2.12) of Clifford et al. (1987). The expression (for $|y| \ge s_N$) is

$$\begin{aligned} P^y[T_N \le t] &= (s_N/|y|) \operatorname{erfc}\left(\frac{|y| - s_N}{\sqrt{2t}}\right) \\ &= \left(\frac{2s_N}{|y|\sqrt{\pi}}\right) \int_{|y|/\sqrt{2t}}^{\infty} \mathrm{e}^{-s^2} \, \mathrm{d}s + \left(\frac{2s_N}{|y|\sqrt{\pi}}\right) \int_{(|y| - s_N)/\sqrt{2t}}^{|y|/\sqrt{2t}} \mathrm{e}^{-s^2} \, \mathrm{d}s \\ &= 2\pi s_N \int_0^t p_s(y) \, \mathrm{d}s + \left(\frac{2s_N}{|y|\sqrt{\pi}}\right) \int_{(|y| - s_N)/\sqrt{2t}}^{|y|/\sqrt{2t}} \mathrm{e}^{-s^2} \, \mathrm{d}s \end{aligned} \tag{7.2}$$

(the last line is the result of routine integration). For $|y| > 2s_N$ (so $|y - s_N| \ge |y|/2$), the second term in the right-hand side of (7.2) is bounded above by

$$\left(\frac{2s_N}{|y|\sqrt{\pi}}\right) s_N (2t)^{-1/2} \exp\{-|y - s_N|^2/(2t)\}$$

$$\le \left(\frac{2s_N^2}{|y|^2\sqrt{\pi}}\right)(|y|^2/(2t))^{1/2}\,\mathrm{e}^{-|y|^2/(8t)}$$

$$\le c(s_N^2/|y|^2)$$

where $c = (2\pi^{-1/2})\sup_{z>0}(z^{1/2}\mathrm{e}^{-z/4})$. Approximating in (7.1) to $P^y[T_N \le t]$ by the first term in the right-hand side of (7.2), we have

$$\begin{aligned}|s_N^{-1} E_N[a(\tilde{Y}_{12}(0)) I_{\{0<T_{12}\le t\}} I_{\{|Y_{12}(0)|\ge 2s_N\}}]\\ - 2\pi\int_{\mathbb{R}^d}\int_{|y|\ge 2s_N} a(\tilde{y})\int_0^t p_s(y)\,\mathrm{d}s\, v(y,\tilde{y})\,\mathrm{d}y\,\mathrm{d}\tilde{y}|\\ \le cs_N\int_{\mathbb{R}^d}\int_{|y|\ge 2s_N} |a(\tilde{y})||y|^{-2} v(y,\tilde{y})\,\mathrm{d}y\,\mathrm{d}\tilde{y}.\end{aligned} \tag{7.3}$$

But

$$\int_{\mathbb{R}^d}\int_{\mathbb{R}^d} |y|^{-2} v(y,\tilde{y})\,\mathrm{d}\tilde{y}\,\mathrm{d}y < \infty, \tag{7.4}$$

(split the integral into integrals over $\{|y|\le 1\}$ and $\{|y|\ge 1\}$, and use the integrability of $v(y,\cdot)$), so that the right-hand side of (7.3) is at most a constant times $\|a\|_\infty s_N$. Also,

$$\begin{aligned}|s_N^{-1} E_N[a(\tilde{Y}_{12}(0)) I_{\{T_{12}\le t\}} I_{\{|Y_{12}(0)|\le 2s_N\}}]|\\ \le s_N^{-1}\|a\|_\infty P_N[|Y_{12}(0)| < 2s_N]\\ \le cs_N^2\|a\|_\infty.\end{aligned} \tag{7.5}$$

Finally, for $t \le \tau$,

$$\begin{aligned}\left|\int_{\mathbb{R}^d}\int_{|y|\le 2s_N} a(\tilde{y})\int_0^t p_s(y)\,\mathrm{d}s\, v(y,\tilde{y})\,\mathrm{d}y\,\mathrm{d}\tilde{y}\right| \le c\,\|a\|_\infty\int_{|y|\le 2s_N}\int_0^\tau p_s(y)\,\mathrm{d}s\,\mathrm{d}y\\ \le c\,\|a\|_\infty\int_{|y|\le 2s_N}|y|^{-1}\,\mathrm{d}y\\ \le c\,\|a\|_\infty s_N^2.\end{aligned} \tag{7.6}$$

Combining (7.3), (7.5) and (7.6) gives us the desired result. □

Lemma 4. *Suppose $d = 3$. Suppose $h: \mathbb{R}_+ \times \mathbb{R}^3 \to [0,\infty)$ is a bounded measurable function such that for some t_1 and t_2, $0 \le t_1 < t_2 < \infty$, $h(t,\tilde{y}) = 0$ for all $\tilde{y}$ unless $t \in [t_1, t_2]$. Suppose there exists $K < \infty$, such that for each $\tilde{y} \in \mathbb{R}^3$, $h(\cdot,\tilde{y})$ is continuously differentiable on (t_1, t_2) and $|h'(\cdot,\tilde{y})|$ is bounded on (t_1,t_2) by K. Then there exists*

$c < \infty$ and N_0 such that for $N \geq N_0$,

$$|s_N^{-1} E_N h(T_{12}, \tilde{Y}_{12}(0)) - 2\pi \int_{\mathbb{R}^d} \int_{\mathbb{R}^d} \int_0^\infty h(t, \tilde{y}) p_t(y) v(y, \tilde{y}) \, dt \, dy \, d\tilde{y}|$$
$$\leq c s_N^{1/2}. \tag{7.7}$$

Proof. Let (M_N) be a sequence chosen so $M_N s_N^{1/2} \to 1$ as $N \to \infty$.

Let $h_N(\cdot, \tilde{y})$ be a function which is zero outside $[t_1, t_2]$ and which is a step function, with M_N equally spaced steps inside the interval $[t_1, t_2]$. (The steps are in the same places for each $\tilde{y}$). Since $h(\cdot, \tilde{y})$ has a uniformly bounded derivative, we may choose h_N so that for some constant c,

$$\|h_N - h\|_\infty \leq c M_N^{-1}. \tag{7.8}$$

We may also arrange for h_N to be jointly measurable. We have

$$E_N |h(T_{12}, \tilde{Y}_{12}(0)) - h_N(T_{12}, \tilde{Y}_{12}(0))| \leq \|h_N - h\|_\infty P_N[T_{12} \leq t_2]$$
$$\leq c M_N^{-1N} s_N \tag{7.9}$$

by (7.8) and Lemma 2.

Also, $h_N(t, \tilde{y})$ is the sum of M_N functions of the form

$$a(\tilde{y}) I_{\{t_3 < t \leq t_4\}}$$

where $a(\cdot)$ is measurable and $t_3 < t_4 \leq t_2$. So by Lemma 3 there are constants c and N_0 such that for $N \geq \mathrm{N}_0$,

$$|s_N^{-1} E_N h_N(T_{12}, \tilde{Y}_{12}(0)) - 2\pi \int_{\mathbb{R}^d} \int_{\mathbb{R}^d} \int_0^\infty h_N(t, \tilde{y}) p_t(y) v(y, \tilde{y}) \, dt \, dy \, d\tilde{y}|$$
$$\leq c M_N s_N. \tag{7.10}$$

Finally,

$$\int_{\mathbb{R}^d} \int_{\mathbb{R}^d} \int_0^\infty |h_N(t, \tilde{y}) - h(t, \tilde{y})| p_t(y) v(y, \tilde{y}) \, dt \, dy \, d\tilde{y}$$
$$\leq c \|h_N - h\|_\infty \int_{\mathbb{R}^d} \int_{\mathbb{R}^d} |y|^{-1} v(y, \tilde{y}) \, dt \, dy \, d\tilde{y}$$
$$\leq c M_N^{-1}, \tag{7.11}$$

since the last integral is finite. Combining (7.9), (7.10) and (7.11) gives us the desired result (7.7). □

Proof of Theorem 4. In view of (3.3), Lemma 1 and Theorem 3, it suffices to prove that for $f \in \mathscr{S}$ and $\tau > 0$,

$$(N^2 s_N/2)^{-1/2} [E_N \phi_N^\tau(f) - \binom{N}{2} s_N \int_0^\tau \int_{\mathbb{R}^d} f(t,x)\,\lambda(t,x)\,\mathrm{d}x\,\mathrm{d}t] \to 0 \quad \text{as } N \to \infty . \tag{7.12}$$

As in the proof of Theorem 1, $E_N \phi_N^\tau(f) = \binom{N}{2} E_N[h(T_{12}, \tilde{Y}_{12}(0))]$, where h is given by

$$h(t,\tilde{y}) = I_{(0,\tau]}(t) \int_{\mathbb{R}^d} f(t,x)\,p_t(x-\tilde{y})\,\mathrm{d}x, \quad (t,\tilde{y}) \in [0,\infty] \times \mathbb{R}^d.$$

By Itô's formula, for $0 < t < \tau$,

$$\left|\frac{\partial}{\partial t} h(t,\tilde{y})\right| = \left|\int_{\mathbb{R}^3} \left[\frac{\partial}{\partial t} + \frac{1}{2}\Delta\right] f(t,x)\,p_t(x-\tilde{y})\,\mathrm{d}x\right| \le \left\|\left(\frac{\partial}{\partial t} + \frac{1}{2}\Delta\right) f\right\|_\infty$$

which is finite, since $f \in \mathscr{S}$.

By Lemma 4, the left-hand side of (7.12) is at most

$$(N^2 s_N/2)^{-1/2} \binom{N}{2} \left| E_N\, h(T_{12}, \tilde{Y}_{12}(0)) - s_N \int_0^\tau \int_{\mathbb{R}^3} f(t,x)\,\lambda(t,x)\,\mathrm{d}x\,\mathrm{d}t \right|$$

$$\le c(N^2 s_N)^{-1/2} N^2 s_N^{3/2}$$

which converges to 0 as $N \to \infty$. □

Proof of Theorem 5. By Theorem 2 and Lemma 1, it suffices to show that for any bounded interval J, contained in $[0,\tau)$ say, we have

$$(N^2 s_N/2)^{-1/2} \left\{ E_N \phi_N^\tau (J \times \mathbb{R}^d) - \binom{N}{2} s_N \int_J \lambda_1(s)\,\mathrm{d}s \right\} \to 0. \tag{7.13}$$

But $E_N \phi_N^\tau(J \times \mathbb{R}^d) = \binom{N}{2} P_N[T_{12} \in J]$, and by Lemma 3 the left-hand side of (7.13) is at most a constant times $(N^2 s_N)^{-1/2} \binom{N}{2} s_N^2$, which converges to zero as $N \to \infty$. □

8. Proof of Theorems 2 and 3 (general case).

Fix $\tau > 0$ throughout this section. We shall study the combinatorics of this proof using the language of graph theory. We shall identify a graph with the set of its edges. A natural random graph on $\{1, 2, \ldots, N\}$ on our probability space Ω_N is obtained by taking its edges to be those $\{i,j\}$ for which $T_{ij} \le \tau$. Divide these edges into two classes, those for which $T_{ij} = 0$ and those for which $T_{ij} > 0$. Since the strictly positive T_{ij} are distinct almost surely, there is a natural ordering on the second class of edges of this graph, determined by the order of the T_{ij}. This graph, together with its subdivision and the ordering, determines the set of $\{i,j\}$ for which particles i and j collide and annihilate before time τ (that is, $T_{ij} = T^k < \tau$ for some $k \leq L$, in the notation of Section 2).

Let $\mathscr{G}^N$ denote the following class of objects. An element G of $\mathscr{G}^N$ is a triple $G = (G_0, G_+, <_G)$, where G_0 and G_+ are graphs on $\{1, 2, \ldots, N\}$, such that the set of edges of G_0 is disjoint from the set of edges of G_+, their union is a connected non-empty graph on some subset of $\{1, 2, \ldots, N\}$, and $<_G$ is a total ordering on the edges of G_+.

We can write G_0 as a set of distinct edges on $\{1, 2, \ldots, N\}$, and G_+ as an ordered sequence of edges on $\{1, 2, \ldots, N\}$, distinct from those in G_0; that is, for $G = (G_0, G_+, <_G) \in \mathscr{G}^N$, we can write

$$G_0 = \{\{i_1, j_1\}, \ldots, \{i_k, j_k\}\}, \tag{8.1}$$

and

$$G_+ = \{\{i_{k+1}, j_{k+1}\}, \ldots, \{i_m, j_m\}\}, \tag{8.2}$$

with

$$\{i_{k+1}, j_{k+1}\} <_G \{i_{k+2}, j_{k+2}\} <_G \cdots <_G \{i_m, j_m\}, \tag{8.3}$$

and $G_0 \cup G_+ = \{\{i_1, j_1\}, \ldots, \{i_m, j_m\}\}$, a connected graph on a subset of $\{1, \ldots, N\}$. Write $\{i, j\} \in G$ if $\{i, j\}$ is an edge of G_0 or of G_+. For G given by (8.1)–(8.3), define the event F_G on Ω_N by

$$F_G = \{T_{i_1 j_1} = T_{i_2 j_2} = \cdots = T_{i_k j_k} = 0\}$$
$$\cap \{0 < T_{i_{k+1} j_{k+1}} < T_{i_{k+2} j_{k+2}} < \cdots < T_{i_m j_m} < \tau\}.$$

If $G, G' \in \mathscr{G}^N$, we shall say G' is an ordered subgraph of G if $G'_0 \subset G_0$, $G'_+ \subset G_+$ and the orderings $<_G$ and $<_{G'}$ on edges of G'_+ coincide. Note that in this case, $F_G \subset F_{G'}$.

Proposition 2. *There is a constant c depending only on d, τ and the initial density function u, such that for every $N > m \geq 1$ and $G = (G_0, G_+, <_G) \in \mathscr{G}^N$, such that $G_0 \cup G_+$ is a tree with m edges,*

$$P_N[F_G] \leq (c s_N)^m \tag{8.4}$$

Proof. For $x_1, \ldots, x_{m+1} \in \mathbb{R}^d$, let $P^N_{x_1, \ldots, x_{m+1}}$ denote probability with respect to N independent Wiener processes denoted $X_1(\cdot), X_2(\cdot), \ldots, X_N(\cdot)$ as before, but now with $X_i(0) = x_i$, $1 \leq i \leq m+1$, and $X_i(0)$, $m+2 \leq i \leq N$, i.i.d. with density $u(\cdot)$ as before. Let $E^N_{x_1, \ldots, x_{m+1}}$ denote the corresponding expectation.

We shall prove that for some constant c_1 depending only on d, if $G_+ \cup G_0$ is a tree on $\{1,2,\dots,m+1\}$, then for $x_1 \in \mathbb{R}^d$,

$$\int_{\mathbb{R}^d} \cdots \int_{\mathbb{R}^d} P^N_{x_1, x_2, \dots, x_{m+1}}[F_G]\,\mathrm{d}x_2 \dots \mathrm{d}x_{m+1} \le (c_1 s_N)^m, \tag{8.5}$$

which implies (8.4) with $c = c_1 \|u\|_\infty$.

We prove (8.5) by induction on m. If $m = 1$, then $G_0 \cup G_+ = \{\{1,2\}\}$. By Lemme 2.1 of Le Gall (1986), with f_d as in that result (f_d is given by (4.6)),

$$s_N^{-1} \int_{\mathbb{R}^d} P^N_{x,y}[F_G]\,\mathrm{d}y \le c_0 \int_{\mathbb{R}^d} f_d(\tau^{-1/2}|y-x|)\,\mathrm{d}y \tag{8.6}$$

where c_0 depends only on d and τ. This implies (8.5) for $m = 1$, when we set c_1 to be the right-hand side of (8.6), which is finite.

Now suppose $m > 1$, and $G = (G_0, G_+, <_G)$, with $G_0 \cup G_+$ an ordered tree on $\{1,2,\dots,m+1\}$. Write G_0 and G_+ as in (8.1)–(8.3).

Consider the case $i_1 \neq 1$ (the case $i_1 = 1$ is tackled by a similar argument to the one below, which we omit). With no loss of generality, assume $i_1 = 2$ and $j_1 = 3$. Also without loss of generality, assume 3 is closer to 1 than 2 is, in the sense that the path from 2 to 1 along G passes through 3 (if this is not true, then 2 is closer to 1 than 3 is; interchange 2 and 3 in the argument below).

By the change of variable $\tilde{x}_2 = x_2 - x_3$, the left side of (8.5) equals

$$\int_{\mathbb{R}^d} \cdots \int_{\mathbb{R}^d} P^N_{x_1, \tilde{x}_2 + x_3, x_3, \dots, x_{m+1}}[F_G]\,\mathrm{d}\tilde{x}_2\,\mathrm{d}x_3 \dots, \mathrm{d}x_{m+1}.$$

By the strong Markov property, this is at most

$$\int_{\mathbb{R}^d} \cdots \int_{\mathbb{R}^d} E^N_{x_1, \tilde{x}_2 + x_3, x_3, \dots, x_{m+1}} I_{\{T_{23} \le \tau\}} P^N_{X_1(T_{23}), \dots, X_{m+1}(T_{23})}[F_{G'}]\,\mathrm{d}\tilde{x}_2\,\mathrm{d}x_3 \dots, \mathrm{d}x_{m+1} \tag{8.7}$$

where $G' = (G'_0, G'_+, <_{G'})$ is the ordered subgraph of G obtained by removal of $\{2,3\}$ from G. One construction of the probability measure $P^N_{x_1, \dots, x_{m+1}}$ is to arrange to have, on a probability space $(\Omega'_N, \mathscr{F}'_N, P'_N)$, a set of N independent d-dimensional Wiener processes $B_i(\cdot)$, $1 < i \le N$, each starting at 0, and a set of $N-(m+1)$ independent $\mathbb{R}^d$-valued random variables $X_i(0)$, $m+1 < i \le N$, with density u; then set

$$X_i(t) = x_i + B_i(t) \qquad (1 \le i \le m+1),$$

$$X_i(t) = X_i(0) + B_i(t) \quad (m+1 < i \le N).$$

With this construction, the expression (8.7) becomes

$$\int_{\mathbb{R}^d} \int_{\mathbb{R}^d} \cdots \int_{\mathbb{R}^d} \int_{\Omega'_N} I_{\{T_{23} \le \tau\}}$$

$$P^N_{x_1 + B_1(T_{23}), \tilde{x}_2 + x_3 + B_2(T_{23}), x_3 + B_3(T_{23}), \dots, x_{m+1} + B_{m+1}(T_{23})}[F_{G'}]\,\mathrm{d}P'_N\,\mathrm{d}\tilde{x}_2\,\mathrm{d}x_3 \dots \mathrm{d}x_{m+1}. \tag{8.8}$$

Since T_{23} depends only on $\tilde{x}_2$ and the Wiener processes $B_i(\cdot)$ $1 \le i \le N$ (in fact, only $B_2(\cdot)$ and $B_3(\cdot)$), we may take the $x_3, \dots, x_{m+1}$ integrations inside the others, so that expression (8.8) is at most

$$\int_{\mathbb{R}^d} \int_{\Omega_N'} I_{\{T_{23} \le \tau\}}$$

$$\times \sup_{y_1, \dots, y_{m+1}} \left\{ \int_{\mathbb{R}^d} \cdots \int_{\mathbb{R}^d} P^N_{y_1, y_2, y_3 + x_3, \dots, y_{m+1} + x_{m+1}} [F_{G'}] \, \mathrm{d}x_3 \dots \mathrm{d}x_{m+1} \right\} \mathrm{d}P_N' \, \mathrm{d}\tilde{x}_2. \quad (8.9)$$

In general, G' splits into two components G^2 and G^3, where 2 is a vertex of G^2 and 3 is a vertex of G^3. By application of the inductive hypothesis to G^2 and then to G^3, the middle line of (8.9) is at most $(c_1 s_N)^{m-1}$, so that expression (8.9) is at most

$$(c_1 s_N)^{m-1} \int_{\mathbb{R}^d} P_N' [T_{23} \le \tau] \, \mathrm{d}\tilde{x}_2,$$

and by (8.6), this is at most $(c_1 s_N)^m$ as desired. □

Let $\mathscr{G}_n^N$ be the set of $G \in \mathscr{G}^N$, such that the graph $G_0 \cup G_+$ has n vertices. Let $\mathscr{G}_n = \bigcup_{N \ge n} \mathscr{G}_n^N$. As a consequence of Proposition 2 we have the following lemma.

Lemma 5. *Under the hypothesis of Theorem 2, there exists n_0 such that*

$$\sum_{G \in \mathscr{G}^N_{n_0+1}} P_N[F_G] \to 0 \quad \text{as } N \to \infty. \quad (8.10)$$

Proof. Take n_0 such that $N^{n_0+1} s_N^{n_0} \to 0$ as $N \to \infty$. This is possible by the hypothesis that $N^{1+\varepsilon} s_N \to 0$ for some $\varepsilon > 0$.

For any $G \in \mathscr{G}^{n_0+1}_{n_0+1}$, G has an ordered subgraph in $\mathscr{G}_{n_0+1}$ which is a tree, so that by Proposition 2,

$$P_N[F_G] \le c^{n_0} s_N^{n_0}.$$

There are only finitely many G in $\mathscr{G}^{n_0+1}_{n_0+1}$, and the number of size $n_0 + 1$ subsets of $\{1, \dots, N\}$ is less than N^{n_0+1}. So for some $c < \infty$,

$$\sum_{G \in \mathscr{G}^N_{n_0+1}} P_N[F_G] \le c s_N^{n_0} N^{n_0+1}$$

and (8.10) follows. □

Suppose $G \in \mathscr{G}^N$. Denote by M_G the event that F_G occurs maximally in the sense that F_G occurs but there is no $G' \in \mathscr{G}^N$, $G' \neq G$, with G an ordered subgraph of G', such that $F_{G'}$ occurs.

Suppose $G \in \mathscr{G}^N$. Then if M_G occurs, for $\{i,j\} \in G$ the question of whether particles i and j annihilate one another at a strictly positive time before τ (that is, $0 < T_{ij} = T^k \leq \tau$ for some k) is fully determined by the structure of G.

Define the function β_G on edges by setting $\beta_G(\{i,j\}) = 1$ if $\{i,j\} \in G$ is such that $0 < T_{ij} = T^k \leq \tau$ for some k whenever M_G occurs. Set $\beta_G(\{i,j\}) = 0$ for all other $\{i,j\}$, including $\{i,j\} \notin G$. For example, if $G = (G_0, G_+, <_G)$, with $G_0 = \{\{1,2\}\}$, $G_+ = \{\{3,4\}, \{2,3\}, \{3,5\}, \{2,6\}\}$ and

$$\{3,4\} <_G \{2,3\} <_G \{3,5\} <_G \{2,6\},$$

then $\beta_G(\{3,4\}) = 1$, and $\beta_G(\{i,j\}) = 0$ for all other $\{i,j\}$.

We have for any function $f \in L^\infty(\mathbb{R}_+ \times \mathbb{R}^d)$, with probability 1

$$\eta_N^\tau(f) = \sum_{G \in \mathscr{G}^N} \sum_{1 \leq i < j \leq N} I_{M_G} \beta_G(\{i,j\}) f(T_{ij}, \tilde{Y}_{ij}(T_{ij})), \tag{8.11}$$

the exceptional event being contained in the event that the $\{T_{ij}, 0 < T_{ij} \leq \tau\}$ are not distinct.

For each $G = (G_0, G_+, <_G) \in \bigcup_{N \geq 2} \mathscr{G}^N$, define the integer valued function γ_G on edges inductively by

$$\gamma_G(\{i,j\}) = \beta_G(\{i,j\}) \quad \text{if } G \in \mathscr{G}_2;$$

$$\gamma_G(\{i,j\}) = \beta_G(\{i,j\}) - \sum_{G'} \gamma_{G'}(\{i,j\}) \quad \text{if } G \in \mathscr{G}_n,\ n > 2,$$

where the last sum is over ordered, connected proper subgraphs G' of G. It follows from the definition that for $1 \leq i < j$,

$$\beta_G(\{i,j\}) = \sum_{G'} \gamma_{G'}(\{i,j\})$$

where the sum is over all connected subgraphs G' of G (including $G' = G$). Also, $\gamma_G(\{i,j\}) = 0$ if $\{i,j\} \notin G$ (proof by induction). By (8.11) we have for $f \in L^\infty(\mathbb{R}_+ \times \mathbb{R}^d)$,

$$\eta_N^\tau(f) = \sum_{G \in \mathscr{G}^N} \sum_{i < j \leq N} I_{F_G} \gamma_G(\{i,j\}) f(T_{ij}, \tilde{Y}_{ij}(T_{ij}))$$

$$= \sum_{G \in \mathscr{G}^N} V_{f,N}(G) \tag{8.12}$$

where we set

$$V_{f,N}(G) := I_{F_G} \sum\sum_{1 \le i < j \le N} \gamma_G(i,j) f(T_{ij}, \tilde{Y}_{ij}(T_{ij})).$$

Let $f \in \mathscr{S}$ or let f be the characteristic function of a set in $\mathscr{R}_{d+1}$. Define the random distribution $\tilde{\phi}_N^\tau$ by

$$\tilde{\phi}_N^\tau(f) := \sum_{n=2}^{n_0} \sum_{G \in \mathscr{G}_n^N} V_{f,N}(G)$$

$$= \sum\sum_{i<j \le N} \sum_{n=2}^{n_0} \sum_{G \in \mathscr{G}_n^N} I_{F_G} \gamma_G(i,j) f(T_{ij}, \tilde{Y}_{ij}(T_{ij})) \tag{8.13}$$

where n_0 is as in Lemma 5. Since every ordered graph in $\mathscr{G}_n^N$, $n > n_0$, has an ordered subgraph in $\mathscr{G}_{n_0+1}^N$, it is immediate from Lemma 5 that $P_N[\tilde{\phi}_N^\tau(f) = \eta_N^\tau(f)] \to 1$ as $N \to \infty$. We shall prove the following results.

Proposition 3. *Under the hypothesis of Theorem* 2, *for* $f \in L^\infty(\mathbb{R}_+ \times \mathbb{R}^d)$,

$$(N^2 s_N/2)^{-1/2} E_N |\eta_N^\tau(f) - \tilde{\phi}_N^\tau(f)| \to 0 \quad \textit{as } N \to \infty.$$

Proposition 4. *Under the hypothesis of Theorem* 2, *for* $f \in \mathscr{S}$ *or* $f = I_R$ *with* $R \in \mathscr{R}_{d+1}$,

$$(N^2 s_N/2)^{-1/2} [\tilde{\phi}_N^\tau(f) - E_N \tilde{\phi}_N^\tau(f)] \xrightarrow{d} W^\tau(f) \quad \textit{as } N \to \infty,$$

where $W^\tau(f)$ *is normally distributed with mean* 0 *and variance*

$$\int_0^\tau \int_{\mathbb{R}^d} f^2(t,x)\, \lambda(t,x)\, \mathrm{d}x\, \mathrm{d}t.$$

By these two results, $\zeta_N^\tau(f)$ converges in law to $W^\tau(f)$. The general cases of Theorems 2 and 3 now follow as in Section 6.

Proof of Proposition 3. We shall show there is a number c depending on n_0 but not on N, such that for large N,

$$|\tilde{\phi}_N^\tau(f) - \eta_N^\tau(f)| < c \, \|f\|_\infty \sum_{G \in \mathscr{G}_{n_0+1}^N} I_{F_G} \tag{8.14}$$

which implies the desired result, by Lemma 5 and the assumption $N^2 s_N \to \infty$. To prove (8.14), first note that the contribution of $\{i,j\}$ to the expression (8.12) for $\eta_N^\tau(f)$ is either 0 or $f(T_{ij}, \tilde{Y}(T_{ij}))$, and has absolute value of at most $\|f\|_\infty$. Also, the contribution of $\{i,j\}$ to the expression (8.13) for $\tilde{\phi}_N^\tau(f)$ has absolute value of at most

$K \|f\|_\infty \sum_G I_{F_G}$, where the sum is over those $G \in \bigcup_{n \le n_0} \mathscr{G}_n^N$ for which $\{i,j\} \in G$, and we set

$$K := \max\{|\gamma_G(\{1,2\})| : G \in \bigcup_{n=2}^{n_0} \mathscr{G}_n\} \tag{8.15}$$

which is finite.

The contribution of edge $\{i,j\}$ to $\eta_N^\tau(f)$ differs from its contribution to $\tilde{\phi}_N^\tau(f)$ only when there exists $G \in \mathscr{G}_{n_0+1}^N$ with $\{i,j\} \in G$, such that F_G occurs. Denoting this event as $H_{\{i,j\}}$, we have by the last two estimates that

$$|\eta_N^\tau(f) - \tilde{\phi}_N^\tau(f)| \le \|f\|_\infty \sum\sum_{i<j\le N} I_{H_{\{i,j\}}} \left(1 + K \sum_{G \in \bigcup_{n \le n_0} \mathscr{G}_n^N : \{i,j\} \in G} I_{F_G}\right). \tag{8.16}$$

Now there exists a number c_2 such that for every $N > n_0$, every $G \in \mathscr{G}_{n_0+1}^N$ has at most c_2 ordered subgraphs in $\bigcup_{n=1}^{n_0} \mathscr{G}_n^N$.

Suppose the event $H_{\{i,j\}}$ occurs. Then for each $G \in \bigcup_{n \le n_0} \mathscr{G}_n^N$ with $\{i,j\} \in G$ such that F_G occurs, G may be extended to some $G' \in \mathscr{G}_{n_0+1}^N$ such that $F_{G'}$ occurs and G is an ordered subgraph of G'. If we do this for each $G \in \bigcup_{n \le n_0} \mathscr{G}_n^N$ with $\{i,j\} \in G$, we cannot obtain the same G' more than c_2 times. Hence,

$$\sum_{G \in \bigcup_{n \le n_0} \mathscr{G}_n^N : \{i,j\} \in G} I_{F_G} \le c_2 \sum_{G' \in \mathscr{G}_{n_0+1}^N : \{i,j\} \in G'} I_{F_{G'}}.$$

whenever $H_{\{i,j\}}$ occurs.

Thus the right-hand side of (8.16) is at most

$$\|f\|_\infty \sum\sum_{i<j\le N} I_{H_{\{i,j\}}} \left(1 + Kc_2 \sum_{G \in \mathscr{G}_{n_0+1}^N : \{i,j\} \in G} I_{F_G}\right), \tag{8.17}$$

and since each $G \in \mathscr{G}_{n_0+1}^N$ has at most $n_0(n_0+1)/2$ edges, there is a constant c such that expression (8.17) is at most

$$c \|f\|_\infty \sum_{G \in \mathscr{G}_{n_0+1}^N} I_{F_G}$$

and (8.14) follows by Lemma 5. □

Proof of Proposition 4. By definition, $\sum_{G \in \mathscr{G}_2^N} V_{f,N}(G) = \phi_N^\tau(f)$. We shall show

$$\mathrm{Var}_N[(N^2 s_N)^{-1/2} \sum_{n=3}^{n_0} \sum_{G \in \mathscr{G}_n^N} V_{f,N}(G)] \to 0 \quad \text{as } N \to \infty, \tag{8.18}$$

which implies the desired result by use of the definition (8.13) of $\tilde{\phi}_N^{\tau}(f)$, Proposition 1, and Billingsley (1968, page 28, Problem 1). The left-hand side of (8.18) equals

$$(N^2 s_N/2)^{-1} \sum_{G',G''} (E[V_{f,N}(G')V_{f,N}(G'')] - E_N V_{f,N}(G') E_N V_{f,N}(G'')) \tag{8.19}$$

where the sum is over G' and G'' in $\bigcup_{n=3}^{n_0} \mathcal{G}_n^N$. The only non-zero terms in the last sum are those for which G' and G'' have at least one vertex in common (since otherwise $V_{f,N}(G')$ and $V_{f,N}(G'')$ are independent). In this case the union of the edges of G' and G'' is a connected graph, denoted G (with no ordering), with between 3 and $(2n_0 - 1)$ vertices. Hence the expression (8.19) equals

$$(N^2 s_N/2)^{-1} \sum_{n=3}^{2n_0-1} \sum_G \sum_{G',G''} \{E_N[V_{f,N}(G)\, V_{f,N}(G')] - E_N V_{f,N}(G') E_N V_{f,N}(G'')\} \tag{8.20}$$

where the second sum is over connected graphs G on size n subsets of $\{1, 2, \ldots, N\}$ and the third sum is over all G' and G'' in $\bigcup_{n=3}^{n_0} \mathcal{G}_n^N$ such that the union of the edges of G'_0, G'_+, G''_0 and G''_+ is G. The number of such pairs (G', G'') is at most some constant, depending on n_0 but not N.

Defining K by (8.15), we have for all G' and G'' in the last sum that

$$E_N |V_{f,N}(G')\, V_{f,N}(G'')| \le (K n_0^2/2)^2 \, \|f\|_\infty^2 \, P_N[E_{G'} \cap E_{G''}] \le c s_N^{n-1}$$

(n being the number of vertices of G) where the last inequality follows from Proposition 2. By the same reasoning, since the sum of the number of vertices in G' and the number of vertices in G'' is at least $n + 1$,

$$E|V_{f,N}(G')|\, E|V_{f,N}(G'')| \le c s_N^{n-1}.$$

Hence, the absolute value of expression (8.20) is at most a constant times

$$(N^2 s_N)^{-1} \sum_{n=3}^{2n_0-1} \sum_G s_N^{n-1}$$

where the sum is over the same class of G as before. The number of such G is at most a constant times N^n, so the last expression is at most a constant times

$$(N^2 s_N)^{-1} \sum_{n=3}^{2n_0-1} (N^n s_N^{n-1})$$

which converges to zero as $N \to \infty$, since by assumption $N s_N \to 0$. This completes the proof of (8.18). □

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