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Renormalization-group study of field-theoretic $A+A\rightarrow\varnothing$

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We study field-theoretic $A+A\rightarrow\varnothing$ in a perturbative renormalization-group framework. We calculate the reacted fraction and it is shown that, in addition to the reaction coupling constant, a second "crossover" parameter must be treated to all orders to obtain meaningful long-time results for spatial dimensions $d\leq 2$. At $d=2$ the result is consistent with known rigorous long-time bounds, but the short-time behavior remains inconsistent (trivial) analogously to the Landau ghost [V. Rivasseau, *From Perturbative to Constructive Renormalization* (Princeton Univ. Press, Princeton, 1991)] of QED and ϕ^4 theory. We argue that this inconsistency is true to all orders in the coupling constants.

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The model [1-4] $A+A\rightarrow\varnothing$ describes an autocatalytic chemical reaction in which diffusing particles in a solvent react with each other and become chemically inert (annihilate). It also provides a simplified description of a number of aggregation processes [5]. More generally, it represents any process involving diffusing localized excitations which annihilate (with a certain probability) on contact. This and related problems, in which interparticle reactions are mediated by fluctuations and their diffusive relaxations, have been the subject of a considerable body of theoretical work [1-4,6-12].

In this Rapid Communication we present a renormalization-group analysis of the $A+A\rightarrow\varnothing$ problem. Despite its status as the most basic of all many-body diffusion-reaction problems, a suitable renormalization calculation of observables such as the reaction rate has not yet been accomplished. We will calculate reaction rates in two-dimensional space (details of these calculations, including results for dimensions below two, will appear elsewhere [13]). The most natural first approach is to develop a theory in terms of the reaction coupling λ_0 alone, i.e., expand in powers of the "bare" λ_0 and then introduce a renormalized λ in the standard fashion. Peliti [1] has shown that, remarkably, the renormalization of the coupling constant may be calculated to all orders. Now after introducing a phenomenological small time scale δ, the bare series for, say, the reaction rate in d-dimensional space involves the dimensionless combination $\lambda_0\delta^{(d-2)/2}$. However, there is another dimensionless combination: $\tilde{X}_0\equiv c_0 t\lambda_0$, where c_0 is the initial concentration. Indeed, on careful scrutiny of the bare series one finds that powers of $\tilde{X}_0$ appear in combination with powers of λ_0. This is a considerable complication since (time being unrenormalized) one has to deal with arbitrarily large values of $\tilde{X}_0$ if one is interested in the long-time behavior; $\tilde{X}_0$ must be treated to *all* orders. In the perturbative renormalization-group calculation which we outline below, we have attempted to calculate to all orders in $\tilde{X}_0$ and to first order in the bare two-body reaction rate λ_0 (that is, general terms of the form $\lambda_0\tilde{X}_0^n$ should be included). From the calculations presented below, such a point of view appears to be consistent at least from the standpoint of removal of divergences. The appearance of a large expansion parameter in this manner occurs analogously in the theory of semidilute polymer solutions [14].

Let us begin by noting some general features of this reacting system. For $d>2$ a "law of mass action" (LMA) holds: the reaction rate is proportional to $c^2(t)$ where $c(t)$ is the concentration of unreacted particles. Thus $c\sim 1/t$. For $d<2$ spatial correlations dictate "diffusion-controlled" behavior since particles explore space compactly [11]; at large enough times a given particle (even if weakly reactive) will have reacted with any "target" in its exploration volume ($\sim t^{d/2}$). Thus in a system of volume V the number of particles varies for large t as $V/t^{d/2}$ (the "empty" volume between surviving particles scales as $t^{d/2}$). We conclude that $c(t)/c_0\sim 1/(c_0t^{d/2})$. To summarize, at long times, for $d>2$ the system is weakly coupled while for $d<2$ the system is strongly coupled at its dimension-dependent fixed point. It seems inevitable that in a field-theoretic treatment such behavior implies the "opposite" short-time behavior, i.e., strong coupling for $d>2$ and weak coupling for $d<2$. One would further anticipate "logarithmic" behavior in the upper critical dimension $d=2$, namely, $c(t)\sim \ln t/t$ for long times. For $A+A\rightarrow \varnothing$ this expectation can actually be proven [8].

Given this phenomenology, it appears that field-theoretic $A+A\rightarrow\varnothing$ exhibits behavior similar to that of ϕ^4 theory, though in certain respects $A+A\rightarrow\varnothing$ is much simpler (i.e., the renormalization is known to all orders [1]). Both theories exhibit a nontrivial infrared fixed point behavior below their upper critical dimensions and asymptotic freedom in the infrared at the critical dimension. Furthermore, we will see that $A+A\rightarrow\varnothing$ suffers from the same ultraviolet problems (i.e., Landau ghost) that attend infrared asymptotic freedom at the critical dimension in ϕ^4 theory. Thus it is not without interest to attempt to carefully examine $A+A\rightarrow\varnothing$ at all times in order to gain insight into more complicated field theories. In fact, resummation to zeroth order in λ_0 and to all orders in $\tilde{X}_0$ reproduces the result of mean-field theory (LMA) which is well behaved at the shortest times; one might hope, therefore, that a proper treatment of $\tilde{X}_0$ could remedy the short-time problems. The persistence of these problems, which we demonstrate below, leads us to consider carefully the two-body analog of $A+A\rightarrow\varnothing$ wherein we find the same short-time inconsistency.

We follow the formalism of Doi [6,7], choosing the minimal sink function $S(x)=\delta(x)$, and we work in $2-\epsilon$ dimensions (we use dimensional regularization). We consider spatially random initial conditions with the total number of particles chosen from a Poisson distribution. We want to calculate $N(t)$, the average number of particles unreacted after a time t which, following Doi, is given by the value of $(c_0V+\partial P/\partial\alpha)e^Q$ at $\alpha=1$. Here $P(\alpha,t)\equiv\sum_{N=0}^{N=\infty}\alpha^N P_N(t)$, $P_N(t)$ is the probability that N particles survive at time t, and

$$Q=\left\langle \alpha\left| T\left[\exp\left(-\int_0^t G_I(t')dt'\right)\right]\right| c_0\right\rangle_c .$$

The subscript c refers to connected diagrams, T is a time-ordering operator, and $\langle\alpha| = \langle 0|\exp(\alpha V^{1/2}a_0)$ and $|c_0\rangle = \exp(c_0V^{1/2}a_0^\dagger)|0\rangle$ are coherent states constructed from the boson creation and annihilation operators, a and $a^\dagger$. Finally, an explicit expression for G_I is given by

$$G_I=\frac{\lambda_0}{2V}\sum_{k,k',q} a_{k+q}^\dagger a_{k'-q}^\dagger a_k a_{k'}-\frac{\lambda_0}{2}\sum_q a_q a_{-q} .$$

At this point we expand the above expressions in terms of λ_0. Doing this, one finds that the expressions are multiplied by factors $\lambda_0^n\tilde{X}_0^m$ where $\tilde{X}_0=c_0t\lambda_0$. $\tilde{X}_0$ here is analogous to the parameter $X_0=v_0cN_0^2$ in the equilibrium theory of semidilute polymer solutions [14] (v_0 is the bare excluded volume parameter). By analogy with semidilute polymer solutions and for the reasons outlined previously we attempt a double expansion in $\tilde{X}_0$ and λ_0. That is, for a given order in λ_0, we attempt to include $\tilde{X}_0$ to all orders. The calculation to zeroth order in λ_0 consists in summing all of the "open diagrams" [6] (see Fig. 1). By brute force, we have been able to do this to third order in $\tilde{X}_0$:

$$N(t)/N_0=1-\tilde{X}_0+\tilde{X}_0^2-\tilde{X}_0^3+\cdots , \tag{1}$$

where $N_0\equiv c_0V$. This is suggestive of the LMA, mean-field result $N(t)/N_0=1/[1+\tilde{X}_0]$ and indeed Doi [6] has an indirect argument to show that this is the case. Next we define a dimensionless coupling constant $\gamma_0\equiv\lambda_0\delta^{\epsilon/2}$ and we introduce $X_0\equiv\gamma_0c_0t$. Performing the renormalization $X_0\rightarrow X$ (of course, to zeroth order this is trivial) and assuming the all-orders LMA form we have

$$\frac{N(t)}{N_0}=\frac{1}{1+X} .$$

We now consult the renormalization group to improve this perturbative result. For $A+A\rightarrow\varnothing$ there is only one parameter to renormalize, namely, γ_0-X_0 is renormalized implicitly, i.e., via γ_0—and we know its renormalization exactly (we shall discuss this later): $\gamma_0=\gamma/(1-\gamma/4\pi\epsilon)$. Therefore we know the β function and renormalization-group equation exactly (although it is, in fact, identical to the result at the one-loop level). Standard arguments [14] then tell us ($d=2$) that $N(t)/$

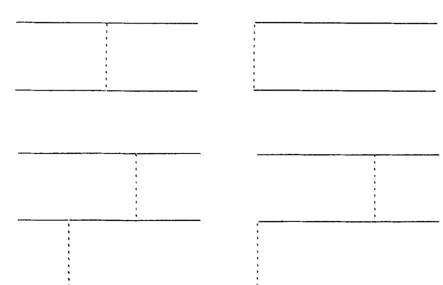

FIG. 1. All "open diagrams" up to order $\tilde{X}_0^2$ are shown. When summed to all orders these diagrams appear to reproduce the mean-field LMA result for $c(t)$.

$N_0=F(X_c,c_0t)$ where $X_c\equiv(1/8\pi)\ln 8\pi t/\delta+1/\gamma$ and F is an arbitrary function which we fix by perturbative calculation. A result consistent with the perturbative result and the renormalization-group equation is

$$\frac{N(t)}{N_0}=\frac{1}{1+c_0t/X_c},$$

whose long-time behavior is well defined and agrees with the known bound [8]. The very shortest-time behavior is bad, however; when t is of the order of δ, X_c becomes negative and consequently there is a time for which $N(t)$ diverges (note that γ is positive). We are thus confronted with the analog of the Landau ghost of quantum electrodynamics and ϕ^4 theory. Higher-order corrections may drastically affect this ghostly behavior. One of the unresolved issues of field theory is whether higher-order corrections mollify this behavior, whether perturbation theory is insufficient to correct the behavior, or whether field theory, itself, is inconsistent (trivial) in these cases.

We next consider the first-order correction to this result, that is we attempt to calculate to order λ_0 and to all orders in $\tilde{X}_0$. Unfortunately this is technically difficult. We were able, however, to carry out this calculation to order $\tilde{X}_0^2\lambda_0$; typical diagrams of this order are shown in Fig. 2. We obtain the following result for the renormalized series:

$$\frac{N(t)}{N_0}=1-X+X^2+\frac{\gamma X}{8\pi}\ln\frac{8\pi t}{\delta}-\frac{\gamma X^2}{4\pi}\ln\frac{8\pi t}{\delta}-\frac{\gamma X}{8\pi}+\frac{\gamma X^2}{8\pi}. \tag{2}$$

[The renormalization $X_0=X(1+\gamma/4\pi\epsilon)$ removes the singularities that arise in the bare series.] A form consistent with the renormalization-group equation and the perturbative result is

$$N(t)/N(0)=1/[1+c_0t/X_c+c_0t/8\pi X_c^2+(c_0t)^2/8\pi X_c^3].$$

The simplest form consistent with the perturbative calculation, the renormalization-group equation, and the known long-time bound is

$$\frac{N(t)}{N_0}=\frac{1}{1+(c_0t/X_c)+(c_0t/8\pi X_c^2)[1/(1-c_0t/X_c)]}. \tag{3}$$

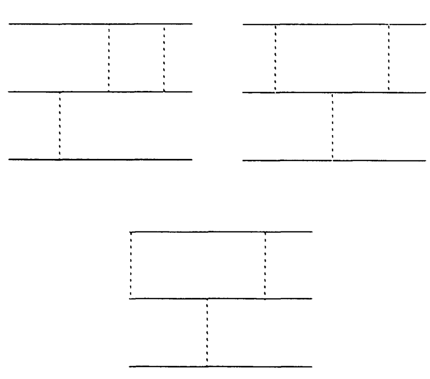

FIG. 2. Examples of order $\tilde{X}_0^2\lambda_0$ diagrams.

This is our final expression for the reaction rate in two dimensions. It is well behaved for large times. The dominant terms for short times are $1-c_0t/X_c-c_0t/X_c^2$ which for "intermediate" times is as one would anticipate on physical grounds; the reacted fraction is small and scales like the volume explored ($\sim t$ in $d=2$) with the expected logarithmic corrections. In fact, if one attempted to renormalize perturbatively in terms of γ only, one would to first order obtain a result close to this short-time form: $N(t)/N_0=1-c_0t/X_c$; clearly such a result is not sensible at large times.

Though our result has good "long" and "intermediate" behavior, it is clear that the behavior is pathological at the shortest times [for example, when $X_c=0$, $N(t)$ vanishes]. The form of the dominant short-time terms suggests that the very-short-time behavior is controlled by the two-body terms, i.e., diagrams with two incoming boson lines (n-body terms have factors of c_0 to the $n-1$ power and consequently t to the $n-1$ power). These diagrams can be summed exactly:

$$\frac{N(p)}{N_0}=\frac{1}{p}-\frac{\lambda_0c_0}{p^2}\left(1+\frac{\lambda_0}{V}\sum_q\frac{1}{p+2q^2}\right)^{-1},$$

where we Laplace transformed $t\rightarrow p$. From this expression one infers that $1/\gamma=1/\gamma_0+1/4\pi\epsilon$, identically to the previous result. Renormalizing this two-body series one finds that the term proportional to $1/p^2$ is positive at very large p. If we assume the small t behavior controls the large p behavior then we conclude that at short times $N(t)/N_0=1+$(positive term). That is, granted seemingly innocuous assumptions, the "all order" very-short-time behavior is nonsense. In order to study more carefully the origin of this problem, we next consider the pure two-body reaction problem. The two-body probability density P obeys $[\partial_t-\Delta_1-\Delta_2-w_0\delta(\mathbf{x}_1-\mathbf{x}_2)]P=0$ (we take a periodic box of size L). The renormalization can be set by looking at the long-time behavior. For a finite system $P(\mathbf{x}_1,\mathbf{x}_2,t\rightarrow\infty)\sim e^{-Et}\tilde{P}(\mathbf{x}_1,\mathbf{x}_2)$ and we can thus consider a d-dimensional Schrödinger equation with a δ-function potential. Such a problem has been previously considered by Berezin and Fadeev [15,16]. These authors proved the existence (independent of dimension) of a nontrivial renormalized solution. (We believe that when one considers the full time dependence this is no longer possible.) The formal argument of Fadeev proceeds as follows. Expanding the Schrödinger equation in terms of a Fourier series one obtains

$$\hat{P}(k)=(-E+k^2)^{-1}(w_0/V)\sum_{k'}\hat{P}(k')$$

and self-consistency demands that $1=(w_0/V)\sum_k[1/(-E+k^2)]$. Since the divergence comes from $|k|\rightarrow\infty$, we rewrite this self-consistency requirement as

$$\frac{1}{w_0}-\frac{1}{V}\sum_k\frac{1}{k^2}=\frac{1}{V}\sum_k\left(\frac{1}{-E+k^2}-\frac{1}{k^2}\right).$$

By choosing $1/w_0=1/w+Z$ where Z is the singular part of $(1/V)\sum_k(1/k^2)$ the divergence is eliminated. We can thus renormalize this problem in any dimension using a suitable regularization (i.e., a momentum cutoff). By using dimensional regularization (about $d=2$) one finds $u_0=u/[1+u/(2\pi\epsilon)]$ where $u_0=w_0\delta^{\epsilon/2}$. The difference in the renormalization compared with $A+A\rightarrow\varnothing$ is only in the choice of name for the parameter.

We next consider the full time dependence inherent in the diffusion equation. A straightforward calculation using Laplace and Fourier transformed variables yields

$$\hat{P}(0,p)=\frac{1}{p}\left[1+\frac{w_0}{Vp}\left(1-\frac{w_0}{V}\sum_k\frac{1}{p+K^2}\right)^{-1}\right]$$

for the Laplace transform of the probability that the particles survive at time t. Renormalizing this expression using the previously determined renormalization of u_0, we find that the term proportional to $1/p^2$ has the form $1/[p^2(A+B\ln p/p_0)]$ where A and B are constants, $A<0$ and $B>0$. Let us assume that large p is determined by short t. Since for large p the expression proportional to $1/p^2$ is positive we see that the correction to 1 at short times is positive, i.e., the probability is greater than 1 at short times. We thus observe the same pathological behavior in this simple model as in $A+A\rightarrow\varnothing$ and we conclude that there is a fundamental inconsistency in both renormalized theories.

In conclusion, we have examined $A+A\rightarrow\varnothing$ in two dimensions and attempted a calculation to order λ_0 where X_0 is included to all orders. Our result, Eq. (3), for the number of surviving particles $N(t)$ is well behaved at long and intermediate times. It would be interesting to see if this form can be distinguished by computer or laboratory experiments from the order λ^0 form. Physically, the necessity to include X_0 to all orders is related to the existence of an important time scale $\bar{t}$, the time to diffuse the initial mean distance between neighbors $(\sim c_0^{-2/d})$. For $t<\bar{t}$ ("intermediate" times) the reacted fraction in general d (provided X_c is small) scales as the volume explored [13]; for $t>\bar{t}$ ("long" times) the surviving fraction scales as the inverse volume explored. This crossover can only be achieved by all-order summation of terms which are powers of the volume explored.

The very-shortest-time behavior, however, is a different story. To a certain extent, the problems of $A+A\rightarrow\varnothing$ have the flavor of the Lee model [17]; however, the many-body version of two-dimensional $A+A\rightarrow\varnothing$ seems to us far away from exactly solvable. The pathological shortest-time behavior of the many-body problem stems from the short-time behavior of the two-body annihilation process and thus in both theories the renormalized series, after improvement by the renormalization group, is nonsensical on these scales. However, the fact that the "intermediate" and "long"-time behaviors are good begs the following question: At what time scales can the renormalized theory be trusted?

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