Comparison of Monte Carlo and analytic results for nearest-available-neighbor pairing in one, two, and three dimensions

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We present the results of large Monte Carlo simulations of nearest-available-neighbor recombination in one, two, and three dimensions of initially random distributions of electrons and holes. The results are compared with analytic findings valid at large and small pair separations. Our results support Eggert's postulated closed formula for the distribution in one dimension. Analytic representations of the data are reported.

The recombination kinetics of an initially random distribution of equal numbers of trapped electrons and holes in a semiconductor remains a fundamental unsolved problem. Physically, such a distribution might be produced by optical illumination if excitonic effects are small, and if, after separation by, for example, diffusive hopping, the carriers become immobile before significant recombination has occurred. The equivalent chemical reaction is $A+B\rightarrow 0$, but we emphasize the absence of classical carrier diffusion. The rates of such reactions, which lead to a wide range of lifetimes, naturally depend on the radiative tunneling mechanism and hence, on the form of the wave functions. At least in principle, this part of the problem is soluble. It turns out that the statistics of the system, and in particular the development of a very nonrandom distribution during the recombination process, is the essential difficulty. For this reason the nearest-available-neighbor (NAN) model has recently attracted attention,[1-5] since it focuses on the latter aspect. Additionally it is sharply and simply defined, has no adjustable parameters, and yields a distribution that does not depend explicitly on time. In it, electron-hole pairs recombine in strict order of their separation, the closest first.

Despite its comparative simplicity, analytic results remain limited to the extremes of small and large electron-hole separations. Suppose that all pairs closer than a distance R have recombined, and that the fraction of the initial population remaining is $Q_D(R)$ in a space of dimension D. By considering clusters of one, two, etc., pairs, Eggert[4] has been able to develop series in R^D for $Q_D(R)$. Exact knowledge of these series is limited at present to six terms in one dimension, and to three terms in two and three dimensions, and so is useful only for small R. However, there were sufficient terms in one dimension for Eggert to propose a plausible closed form for $Q_1(R)$. We have given elsewhere[3] a simple scaling argument based on the stochastic nature of charge fluctuations in the original distribution leading to the prediction that as $R\rightarrow\infty$, $Q_D(R)\sim R^{-D/2}$.

Because of the limitations of known analytic methods, we have previously used a Monte Carlo approach[5] to test the asymptotic prediction which was at that time in doubt, and to examine the region between the limits $R\rightarrow 0$ and $R\rightarrow\infty$ for the three-dimensional case. In the present paper we extend the Monte Carlo results to one and two dimensions with significantly better statistics than those of Eggert,[2] and compare our results with the corresponding analytic predictions. We have also obtained semiempirical analytic fits to the data in two and three dimensions. These fits conform to what is presently known about the distributions at both small and large separations and serve to describe the empirical data over the difficult intervening region.

The Monte Carlo simulations were made with the mutual nearest-neighbor algorithm described before.[5] The initial pair numbers in each run in one, two, and three dimensions were 1.6×10^4, 5×10^5, and 1.6×10^6, respectively. In comparison, Eggert's earlier simulations[2] used 1×10^4 in one dimension and only 5×10^3 in two and three dimensions. Large numbers are essential in establishing the approach to the asymptotic limit for $D>1$. However, for $D=1$ the asymptote was in little doubt and our main concern was to obtain statistics good enough to test Eggert's proposed closed form in the uncertain transition region. To achieve this we performed 248 runs of 16383 initial pairs, employing more than 4×10^6 pairs overall. In two and three dimensions, three independent, much larger runs were made to confirm the asymptote as well as the transition region. Because of our interest in the asymptotic power law, and because of the sensitivity of the differential, we present the results in Figs. 1-3 in the form of the logarithmic slope $G_D(V)=d[\ln Q_D(V)]/d(\ln V)$ vs $\ln V$. V is the dimensionless volume in the appropriate space, i.e., $V=v_D(R/L)^D$ with L the initial pair density to the $(-1/D)$ power and v_D the volume of the unit D sphere.

The results of the simulations are shown in Figs. 1-3. In all three cases $G_D(V)$ is initially 0 at small V and at large V is consistent with the predicted asymptotic values of $-\frac{1}{2}$ within the statistical errors. We now regard this prediction as well established for three and fewer dimensions.

Together with the one-dimensional Monte Carlo results shown in Fig. 1, we have also included Eggert's analytic results.[4] The solid line is the function

$$G_1(V)=-[1-\exp(-2V)]/2\,, \tag{1}$$

which he guessed by extrapolating the calculated form of the first six terms of his series. The agreement is clearly

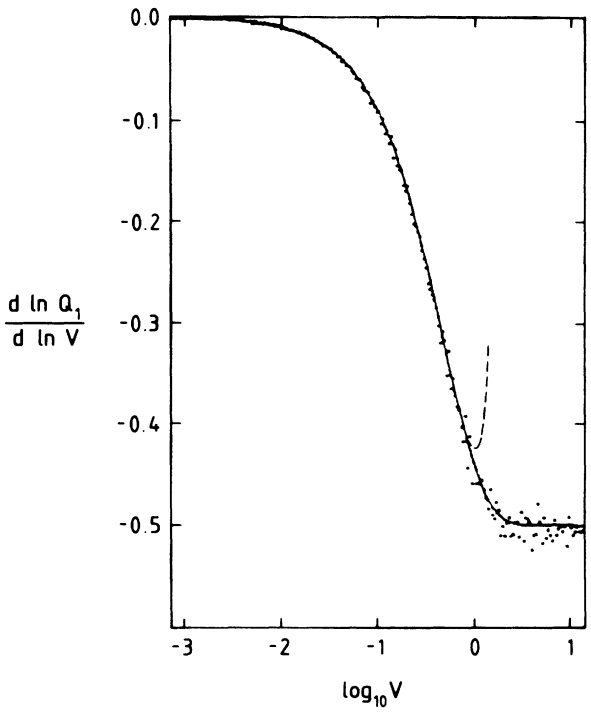

FIG. 1. Monte Carlo results for one dimension averaged over 248 runs each with 16383 initial pairs. Solid line shows Eq. (1), dashed line, Eq. (2).

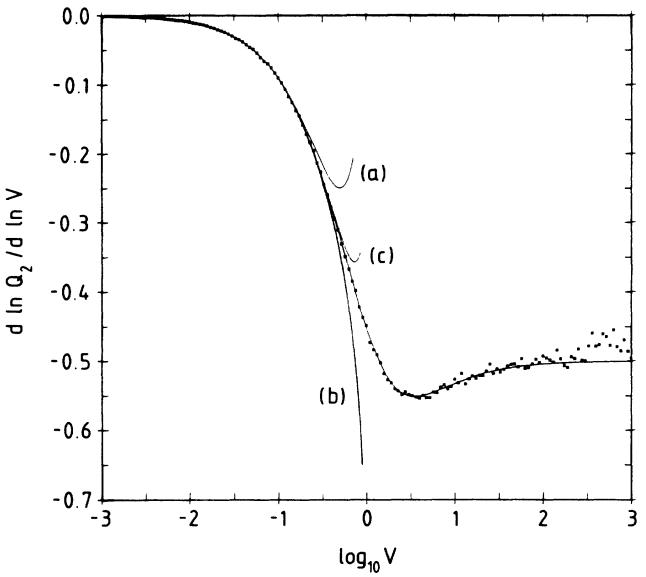

FIG. 2. Monte Carlo results for two dimensions averaged over three runs each with 5×10^5 initial pairs. The solid line is the fit to the data given by Eq. (7). Curve *a* shows the first two terms of Eq. (5); curve *b* includes the third term, the first to include NAN effects. Curve *c* includes our rough estimate of the fourth term, $0.49V^4$.

very good, and lends considerable weight to his proposal. For comparison we also show (dashed line) the first six terms alone,

$$G_1(V)\simeq -V+V^2-\tfrac{2}{3}V^3+\tfrac{1}{3}V^4-\tfrac{2}{15}V^5+\tfrac{2}{45}V^6, \quad (2)$$

since these are certainly correct. $G_1(V)$ appears to approach its asymptotic value without overshoot, though because of the statistical noise we cannot rule such behavior out entirely. The steepest slope seems unlikely to exceed 0.5 by more than 0.01.

Figures 2 and 3 show the results of the two- and three-dimensional simulations. The most striking difference in these higher dimensions is the overshoot following $V=1$. We have previously noted[5] how in three dimensions this effect can produce a misleading asymptotic slope unless the initial population is large enough. The overshoot is deeper in three dimensions than in two, and we speculate that this trend will continue into higher dimensions with increasing freedom from the pairing constraint. In two dimensions the greatest slope is -0.55 and in three dimensions it is -0.62. When considering the approach of G_2 and G_3 towards the predicted asymptote of -0.5, it is essential to be aware that in each case the data represent only three (very large) independent simulations. Towards the end of each run, as R approaches the dimension of the periodic unit cell employed, behavior characteristic of the particular initial pair distribution becomes more and more apparent; see, for example, Fig. 3 of Ref. 5 and Figs. 3 and 4 of Ref. 2. The points at high V in Figs. 2 and 3 of the present work are scattered randomly about an erroneous trend specific to the average of those of the three initially chosen distributions. It so happened that the systematic trends tended to cancel for the three runs of G_3, giving an average close to -0.5, while for G_2 all three broke away upwards; thus for $V>100$, G_2 seems to increase beyond -0.5.

Figures 2 and 3 also include results of Eggert's cluster

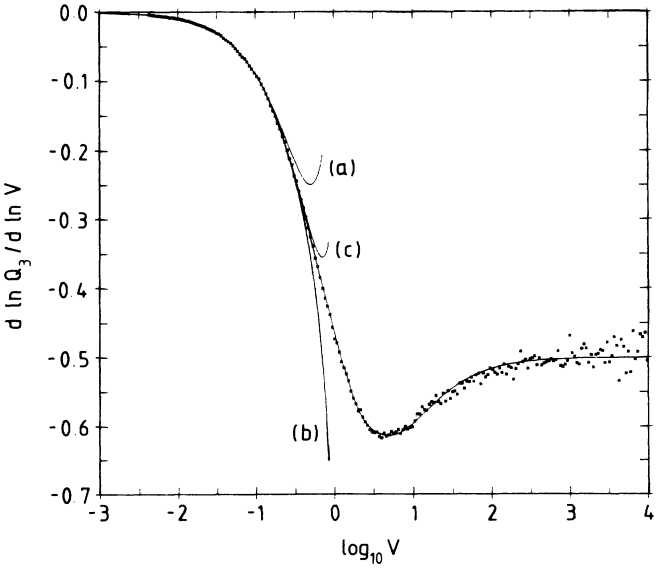

FIG. 3. Monte Carlo results for three dimensions averaged over three runs each with 1.6×10^6 initial pairs. The solid line is the fit to the data given by Eq. (8). Curve *a* shows the first two terms of Eq. (6), curve *b* includes the third term, and curve *c* includes our rough estimate of the fourth, $0.59V^4$.

calculation which is at present limited in two and three dimensions to terms in V^3. While it is, in principle, possible to calculate higher terms, the labor involved increases very rapidly. When we compared the Monte Carlo results with Eggert's published coefficients, we became aware of an error in both two and three dimensions. This was confirmed when the cluster calculations were repeated both by ourselves and by Eggert.[6] The corrected results are

$$Q_2(V) = 1 - V + V^2 - 0.9233966V^3 \tag{3}$$

and

$$Q_3(V) = 1 - V + V^2 - \tfrac{298}{315}V^3 . \tag{4}$$

The first three terms are independent of the dimensionality, and only the fourth reveals NAN effects. Its coefficient becomes closer to -1 as the dimensionality increases. In terms of $G_D(V)$ we have

$$G_2(V) = -V + V^2 - 0.7701898V^3 + \cdots \tag{5}$$

and

$$G_3(V) = -V + V^2 - \tfrac{88}{105}V^3 + \cdots . \tag{6}$$

In Figs. 2 and 3 curve *a* shows the first two of these terms, i.e., the purely non NAN contribution and curve *b* shows all three known terms.

In order to render our numerical results more readily available for application, we have fitted the data for two and three dimensions to simple algebraic expressions which satisfy the asymptotic behavior and agree with Eggert's terms of Eqs. (5) and (6) at small V. The results are shown as solid lines. For two dimensions our fitting function is

$$G_2(V) \simeq (-V + V^2 - 0.77019V^3 + 0.490V^4 - 0.266V^5 + 0.142V^6 - 0.0446V^7 - \tfrac{1}{20}V^8)/(1 + \tfrac{1}{10}V^8) , \tag{7}$$

and in three dimensions

$$G_3(V) \simeq (-V + V^2 - \tfrac{88}{105}V^3 + 0.5915V^4 - 0.3014V^5 + 0.0981V^6 - 0.0169V^7 - \tfrac{1}{200}V^8)/(1 + \tfrac{1}{100}V^8) . \tag{8}$$

It cannot be too strongly stressed that the coefficients beyond the cubic term are progressively underdetermined and should not be regarded as reliable indicators of the true series expansion. The fit is particularly insensitive to the coefficients of V^8 whose function is to enforce the -0.5 asymptote. The purpose of these expressions is merely to give convenient analytic expression to our numerical data.

The agreement with the simulation is worst between the peak of the overshoot and the asymptotic region, but is never more than 0.01 from the mean of the data. Equation (8) is an alternative to our earlier fit of $Q_3(R)$ to the Monte Carlo data,[5] but is an improvement in that it behaves more correctly, i.e., as Eq. (6) at low V; the cubic term in Eq. (4) was not known to us at the time of the earlier work.

From our fits to the three-dimensional data [Eq. (8)] we estimate that the next term in Eq. (6) is $(0.60 \pm 0.05)V^4$. The fit to the two-dimensional data of Eq. (7) leads to a less well-defined fourth term $(0.5 \pm 0.1)V^4$ in Eq. (5). These are crude estimates but may serve to provide a rough check for any future analytic calculation. Curve *c* in Figs. 2 and 3 shows the result of including these estimates of the fourth terms for two and three dimensions. The important point this makes is that many cluster terms would be needed to significantly improve the series description towards higher V. Bearing in mind the rapidly increasing difficulty of evaluating the multidimensional integrals of the later terms, we feel that it is unlikely that further significant progress will be made in this way.

The Monte Carlo results reported here confirm our power-law prediction for large V and Eggert's series for small V in three and fewer dimensions. They also seem to confirm Eggert's suggested $G_1(V)$ [Eq. (1)]. In addition, they provide an accurate numerical representation of the analytically intractable two- and three-dimensional cases of NAN recombination.

[1] D. J. Dunstan, Philos. Mag. B **46**, 579 (1982); Physica **117-118**, 902 (1983).
[2] J. R. Eggert, Phys. Rev. B **29**, 6664 (1984).
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[4] J. R. Eggert, Phys. Rev. B **37**, 4051 (1988).
[5] J. E. L. Bishop and T. M. Searle, Philos. Mag. B **57**, 329 (1988).
[6] J. R. Eggert (private communication).