Recombination in disordered semiconductors: The nearest-available-neighbor distribution

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(Received 7 November 1986; revised manuscript received 21 October 1987)

The distant-pair model describes electron-hole recombination in a disordered semiconductor. An analytic method is presented for the derivation of the power series of the distribution of recombination distances in a dilute distant-pair system. Results are presented for the one-, two-, and three-dimensional systems, and show that previous approximate methods underestimate the density of metastable carriers. An exact sum of the power series is proposed for the one-dimensional case which reproduces results of previous Monte Carlo studies.

I. DISTANT-PAIR MODEL

The distant-pair model describes electron-hole recombination in crystalline[1] and amorphous[2] semiconductors. The model uses as its initial state a distribution of equal numbers of electrons and holes, localized at fixed random positions in space, and interacting only by tunneling recombination. All competing processes, such as thermal detrapping, are excluded from the model. The problem can be examined in two limiting cases. If the average intercarrier distance is large compared with the tunneling range, then because the tunneling rate τ^{-1}, often expressed as

$$\tau^{-1}(r)=\tau_0^{-1}\exp(-\alpha r)\ , \tag{1}$$

is such a strongly decreasing function of tunneling distance, the carriers which are closest to each other will necessarily recombine first. As recombination proceeds, the average intercarrier distance will increase, and the tunneling rate will decrease rapidly. The decay is therefore strongly nonexponential. The problem is completely deterministic, being given by the particular initial distribution of immobile carriers, and by the expression for the tunneling rate. The distribution of recombination distances is determined by the statistics of the pairing of random positions in space. The time dependence of the population density is then simply calculated from the recombination distance distribution and the tunneling rate dependence on recombination distance.

If on the other hand, the carrier density is sufficiently large, then the carriers will not necessarily recombine with the closest partner, but can easily tunnel much further away. In this case, the form of the dependence of the tunneling rate on the recombination distance becomes important, and the time dependence of the recombination must be calculated probabilistically. The pair separation distribution of the dilute case would no longer apply. In this paper, we discuss only the dilute case.

One might think[3] that the distribution of recombination distances for the dilute distant-pair system is just the nearest-neighbor (NN) distribution. For an n-dimensional system this is given by[4]

$$P_{\mathrm{NN},n}(r)=\rho n v_n r^{n-1}\exp(-\rho v_n r^n)\ , \tag{2}$$

where ρ is the density of carriers and v_n is the volume of the unit n-dimensional sphere,

$$v_n=\frac{\pi^{n/2}}{\Gamma(n/2+1)}\ . \tag{3}$$

This distribution is not appropriate, however, because the nearest-neighbor relation that it describes is not commutative, i.e., a carrier is not necessarily the nearest neighbor of its nearest neighbor. Recombination, however, is commutative, for if electron A recombines with hole B, then hole B necessarily recombines with electron A. The nearest-neighbor distribution therefore cannot describe the commutative recombination process.

The correct distribution[2] for the description of distant-pair recombination is the nearest-available-neighbor (NAN) distribution, $P_{\mathrm{NAN}}(r)$. In this pairing scheme, each carrier must recombine with the nearest carrier that is available to it. The definition of availability can be cast in several ways, each of which implies a method for finding the NAN of a particular carrier. One definition of availability reflects the time dependence of the physical process which is being modeled: a carrier A is available at distance r if it still exists at a time $\tau(r)$. This definition is useful for the Monte Carlo calculation of $P_{\mathrm{NAN}}(r)$, but unnecessarily brings in the time dependence of the recombination to describe a spatial relation between carriers.

A second definition of availability, and one which will be used for the calculations in the next section, is a recursive one: a carrier A is available to a carrier of opposite sign D at distance r if there is no carrier of the opposite sign B at distance $r'<r$, which is itself available at distance r'. The recursion reflects the competition for the nearest available neighbor involved in the NAN pairing scheme.

A third definition of availability is embodied in another algorithm for the Monte Carlo calculation of the NAN recombination distance distribution, recently performed by Bishop and Searle.[5] In this calculation, any carrier pair which is mutually nearest neighbor is removed, and its separation is noted. As carrier pairs are removed, other carrier pairs which previously were not mutually nearest neighbors will become so, and thus become candidates for removal. It makes no difference what order the

carriers are removed; time is not directly modeled. This process is essentially the recursive definition run backwards.

Regardless of how one considers the definition of availability, the NAN pairing process is unique when the carriers are positioned at random and equidistance is not a problem. It is easy to see that for sufficiently small r, the NAN competition is unimportant, and one may write

$$P_{\mathrm{NAN},n}(r)\approx\rho n v_n r^{n-1} , \tag{4}$$

which just expresses how many carriers exist at distance r in n-dimensional space.

Several examples of NAN pairing are shown in Fig. 1. In Fig. 1(a), carrier A is available to carrier D of opposite sign at distance r, as no other carriers are nearby. In Fig. 1(b), carrier A is no longer available at distance r, because carrier B is paired with A at distance $r'<r$. In Fig. 1(c), carrier A is now available at distance r, because, although B exists at distance $r'<r$, it is paired with carrier C at distance $b<r'$, and is thus not available at distance r'. In Fig. 1(d) is depicted a group of six NAN pairs, the pairing indicated by lines, and the relative order of recombination distances indicated by numbers. Verification of the correctness of the pairing is left to the reader.

An ansatz used by Dunstan[2] to derive an analytic form $P_{D,n}(r)$ for the NAN distribution is based on standard geometric probability. In this method, generalized elsewhere[6] to n dimensions, the probability of a carrier A finding a NAN at distance r is the product of three conditional probabilities: the probability $\rho n v_n r^n$ that there is a carrier B of opposite sign at r, the chance $[1-\int_0^r P_{D,n}(r')dr']$ that A has not found a NAN nearer than r (i.e., that A is available at distance r), and a similar chance that B in turn has not found a NAN nearer than r. Thus P_D is written

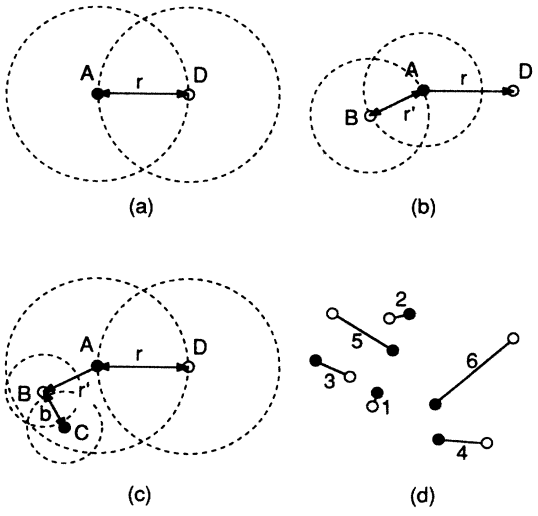


FIG. 1. NAN pairing schemes. (a) Carriers A and D are NAN's. (b) Carriers A and D are not NAN's because of the existence of B. (c) Carriers A and D are NAN's because B is paired with C. (d) A group of six NAN pairs numbered in order of increasing pair separation.

$$P_{D,n}(r)=\rho n v_n r^{n-1}\left[1-\int_0^r P_{D,n}(r')dr'\right]^2 . \tag{5}$$

This equation can be solved by elementary methods,[6] and defining in general the complementary cumulative distribution function

$$Q(r)=\int_r^\infty P(r')dr' , \tag{6}$$

we get

$$Q_{D,n}(x)=\frac{1}{1+v_n x^n} , \tag{7}$$

where $x=\rho^{1/n}r$ is the normalized pair separation. For small x, this result predicts the power series

$$Q_{D,n}(x)=1-v_n x^n+v_n^2 x^{2n}-v_n^3 x^{3n}+\cdots , \tag{8}$$

whereas for large x it gives

$$Q_{D,n}(x)\sim v_n^{-1}x^{-n} . \tag{9}$$

The one-dimensional case of this relation is shown in Fig. 2 as a dashed line.

Unfortunately, the Dunstan ansatz for the NAN distribution has been shown to be based on the inaccurate approximation of uncorrelated conditional probabilities, and deviates significantly from results of Monte Carlo analyses.[6] The Monte Carlo results in one dimension are reproduced in Fig. 2 as a dotted line. Another analytic method for the determination of the NAN distribution must be sought.

An estimate of the asymptotic dependence of the NAN distribution has been proposed by Searle and Bishop,[7] who have hypothesized that the average density of remaining carriers in a distant-pair system should be determined by the large-scale fluctuations from neutrality of the original system. These fluctuations are proportional to the square root of the expected number of carriers in a chosen volume, or if normalized to the original density, to the inverse square root, implying that the asymptotic

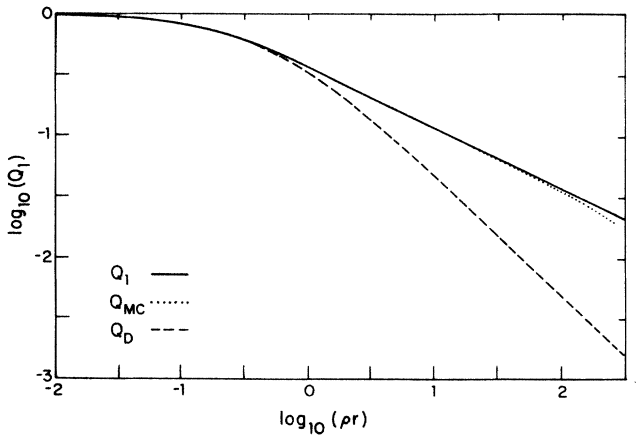


FIG. 2. NAN distributions in one dimension. Dunstan approximation (dashed), Monte Carlo calculation (dotted), and proposed exact sum (solid).

behavior of the NAN distribution should be $Q(r)\sim r^{-n/2}$ for large r. This approach succeeds in reproducing the exponent of the power law dependence of the Monte Carlo NAN distribution relatively well in one,[6] two,[6] and three[5] dimensions in the most extensive calculations made.

A second method for the investigation of the distant-pair model is the direct calculation of the time decay of the recombining system. In this paper we introduce the kinetic analysis only in order to help find the NAN distribution for the dilute distant-pair model. The decay of the distant-pair system was treated in the Hartree approximation by Hoogenstraaten[8] and by Thomas *et al.*[1] The Hartree approximation is expected to be valid only at short times, where the carrier populations are virtually undepleted, and thus should not be expected to provide accurate results for the NAN distribution at large recombination distances.

Another calculation of the distant-pair time decay was given by Dunstan,[2] who wrote the probability of a carrier at the origin of recombining at time t with another carrier of opposite valence at distance r as

$$P_{D,n}(r,t)=nv_n r^{n-1}\rho Q_{D,n}^2(t)\tau^{-1}(r)\exp[-t/\tau(r)] . \quad (10)$$

This expression, as Dunstan's ansatz for $P_{D,n}(r)$, is composed of the product of various probabilities: the chance $nv_n r^{n-1}\rho$ of finding a carrier at distance r, the chance $Q_{D,n}(t)$ that this carrier has not yet recombined by the time t, the chance $Q_{D,n}(t)$ that the carrier at the origin has also not yet recombined, and the recombination rate between two carriers $\tau^{-1}(r)\exp[-t/\tau(r)]$, where $\tau(r)$ is the inverse recombination rate given in Eq. (1). This ansatz may be solved for the time dependence of the carrier density $Q(t)=\int_t^\infty dt'\int_0^\infty dr\,P(r,t')$ to give

$$Q_{D,n}(\theta)=\left[1+\frac{v_n\rho}{\alpha^n}\theta I_n(\theta)\right]^{-1} , \quad (11)$$

where $\theta=t/\tau$. The function $I_n(\theta)$ is given by the integral

$$I_n(\theta)=\int_0^\infty z^n e^{-z}\exp(-\theta e^{-z})dz . \quad (12)$$

Some of the properties of this function are discussed in the Appendix.

In addition to the assumption of independent conditional probabilities, this ansatz uses a time-dependent expression for the recombination rate between two carriers, in addition to the explicit inclusion of the survival probabilities of each of the carriers. It has been suggested[9] that the inclusion of the additional time dependence of the recombination rate expresses a correlation between the survival factors of the two carriers under consideration. No derivation for this correlation has been given. Furthermore, Monte Carlo calculations of the time decay of the distant-pair system in one dimension show significant deviations from the behavior predicted by Eq. (11).

Despite these objects, it is interesting to consider a comparison of the two Dunstan ansatzes, Eqs. (5) and (10). One would expect that

$$\int_0^\infty P_{D,n}(r,t)dt=P_{D,n}(r) \quad (13)$$

in the limit of low density (or equivalently short tunneling range). If we work with the quantity $Q(r)$ as defined previously, it is in fact possible to test this relation. We write

$$Q_{E,n}(r)=\int_0^\infty dt\int_r^\infty dr'\,P_{D,n}(r',t) \quad (14)$$

as the definition of the recombination distance distribution derived from the Dunstan ansatz for the distant-pair decay. This becomes

$$Q_{E,n}(r)=\frac{nv_n\rho}{\alpha^n}\int_0^\infty Q_{D,n}^2(\theta)I_{n-1}(\theta,z)d\theta \quad (15)$$

$$=\sum_{m=0}^{\infty} Q_{n,m}r^m , \quad (16)$$

where

$$Q_{n,m}=\frac{nv_n\rho}{\alpha^n}\int_0^\infty Q_{D,n}^2(\theta)\frac{\alpha^m}{m!}\frac{\partial^m}{\partial z^m}I_{n-1}(\theta,z)d\theta\Big|_{z=0} \quad (17)$$

and $z=\alpha r$. The incomplete decay functions $I_n(\theta,z)$ are discussed in the Appendix. Because $Q_{D,n}(\theta)$ appears in this expression as a squared term, the coefficients $Q_{n,m}$ are difficult to evaluate, but a few can be found exactly in the limit as α goes to infinity:

$$Q_{n,m}=0 \text{ if } m\neq pn,\ p \text{ integer} , \quad (18)$$

$$Q_{E,n}(x)=1-v_n x^n+\tfrac{1}{2}v_n^2 x^{2n}-\cdots , \quad (19)$$

where $x^n=\rho r^n$. The coefficient of the x^{2n} term disagrees with that in the expansion for $Q_{D,n}(x)$ in Eq. (8) by a factor of 2. This term is determined by the contribution of the nearest neighbors, and does not reflect the competition of the NAN pairing scheme. On this level of sophistication, one would expect that the NAN distribution should look like the NN distribution until competition starts to take place. This intuitive approach will be justified below. In this case, it is expected that the expression for $Q_{E,n}(r)$ is the one in error. The second Dunstan ansatz does not correctly account for nearest-neighbor interactions.

It is possible to compare the results of the two Monte Carlo calculations as well. Searle and Bishop[7] have used an analytic form fitted to the Monte Carlo $P_{\mathrm{NAN},1}(r)$ and have calculated from it the time decay of a one-dimensional distant-pair system. In their work, the result coincides reasonably well with the direct Monte Carlo result for the time decay.

II. PERTURBATION EXPANSION METHOD

It has been shown that $P_{D,n}(r)$ and $P_{D,n}(r,t)$ do not agree with Monte Carlo results or the Searle and Bishop prediction, and do not agree with each other in their prediction of the distant-pair recombination distance distri-

bution. It would be desirable to find a way to calculate the NAN distribution analytically, to amplify on the Monte Carlo results. To develop such a method for the NAN problem is difficult, for it must account for the infinitude of pairing competition inherent in the distant-pair system. We will now discuss a method which is able to provide a power series for the NAN distribution.

The basic approach is to use the recursive definition of availability given in Sec. I, and to approximate the NAN distribution by considering successively deeper levels of recursion. To do so, we introduce the concept of an isolated cluster of carriers about a central carrier pair. An isolated cluster is, loosely speaking, composed of all the carriers which have any causative effect on the recombination of the central carrier pair. These are all the carriers which are considered when using the recursive definition of availability given in Sec. I. For any central carrier pair, the isolated cluster is uniquely defined by the positions of all the carriers in the ensemble. Associated with any particular isolated cluster is a set of spheres about each paired carrier in the cluster. Each sphere of influence has the valence of the carrier about which it is centered (i.e., positive or negative). The sphere of influence of a carrier A paired with another carrier B at distance r is the sphere of radius r centered at A, and has the valence of A. The spheres of influence of the paired carriers are depicted in Fig. 1. Within a sphere of influence of valence A and radius r, there can be no carrier of valence B at a radius $a<r$, unless that carrier is paired at a distance $b<a$. The spheres of influence determine when carriers are part of an isolated cluster. In Fig. 1(c), for example, if the carrier pair AD is being considered as the central carrier pair, the isolated cluster contains carriers A, B, C, and D, whereas the isolated cluster about the central carrier pair BC contains only the carriers B and C.

We may now define an isolated cluster to be a finite group of carriers paired off in the NAN scheme, where at least one carrier from each carrier pair except the central carrier pair is in the sphere of influence of another carrier of the opposite valence in the cluster. An isolated cluster must include by definition an equal number of electrons and holes. If there were an unequal number, then after the pairing scheme there would be one or more carriers of the same valence left. Because of the arguments in the previous paragraph, these remaining carriers then cannot lie in any sphere of influence and thus do not belong to the isolated cluster. As one considers the contributions of isolated clusters containing successively more carrier pairs, more of the possible pairing schemes will be included, and the true nearest-available-neighbor distribution will be more closely approximated.

We form the probability of an isolated cluster with a finite number of carrier pairs, and therefore its contribution to the complete distribution of P_{NAN}, by writing the probability for the cluster with a particular set of carrier positions, and averaging over the carrier positions, holding the separation r of the central carrier pair constant, and keeping one of the central pair at the origin. In view of the known density scaling law for P_{NAN}, we can use the scaled positions $\mathbf{x}=\rho^{1/n}\mathbf{r}$ in n dimensions, and then need not explicitly include the carrier density. Let V_A and V_B be the total nonoverlapping volumes of spheres of influence of valence A and B (electrons and holes), respectively. Then the probability of a cluster is

$$P_{\mathrm{cluster}}=\exp(-V_A-V_B)\ . \tag{20}$$

This is to be averaged over the carrier positions that give rise to the particular pairing.

An infinite number of types of isolated clusters is required to calculate P_{NAN} to arbitrary accuracy. One can learn something from choosing a finite number of cluster types, however. Because a cluster of $2N$ carriers is averaged over $(2N-1)$ positions, holding one distance constant, the contribution of a cluster must scale at least as $x^{n(2N-1)-1}$. Thus by taking successively more carriers into consideration, a power series for P_{NAN} can be constructed.

Consider first the case $N=1$, or just one carrier pair in the isolated cluster. Here the volumes of the spheres of influence are easy to write down, they are $V_{A,B}=v_n x^n$. Averaging over the angular position of the carrier at distance r gives

$$P_{n,1}(x)=nv_n x^{n-1}\exp(-2v_n x^n) \tag{21}$$

for the contribution of the one-pair isolated clusters in n dimensions. This is just the set of carriers which are the nearest neighbor of their nearest neighbors. These account for half of all recombinations in the NAN model. To this approximation, the power series for the NAN distribution function is thus

$$P_{\mathrm{NAN},n}(x)\approx nv_n x^{n-1}-2nv_n^2 x^{2n-1}\ . \tag{22}$$

So far the expansion accounts only for nearest-neighbor effects, not for any NAN competition for availability. This is already sufficient to exclude the recombination distance distribution calculated from $P_{D,n}(r,t)$, however, and it justifies the intuitive approach given in Sec. I.

It is possible to write down the expressions for the N-pair contributions to the NAN distribution, but the complete integrals are analytically intractable except in one dimension. In one dimension, it is possible to construct and solve the terms for an arbitrary number of pairs in an isolated cluster. For calculational purposes, the N-pair contributions are separated into particular pairing configurations. In Tables I and II is presented a list of pairing configurations with one-, two-, and three-pair

TABLE I. Summary of NAN isolated clusters: one- and two-pair configurations.

Pairs	Configuration	Notation	Density (ppm)
1		$P_{1\text{-}1}$	500 000
2		$P_{2\text{-}1}$	35 714
2		$P_{2\text{-}2}$	41 666
2		$P_{2\text{-}3}$	10 417
2		$P_{2\text{-}4}$	31 250

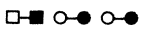
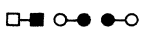
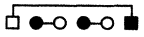
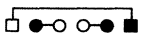
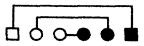
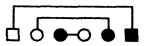
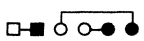
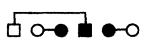
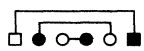
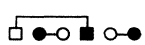
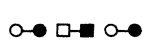
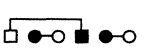
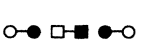
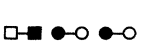
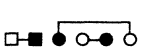
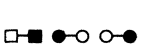
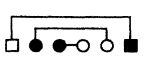
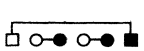
clusters in one dimension. The number of pairs, the configuration, notation, and density for each NAN configuration are listed. Each configuration consists of a placement of the pairs in the one-dimensional space in a particular order. The symbols in the schematic configuration diagrams indicate the carrier type: open and filled symbols indicate the two valences, and the square symbols indicate the central carrier pair. The lines indicate the NAN pairing. The notation is chosen in arbitrary order. The density represents the number of carriers expected to be paired in the manner of the central carrier in each configuration. Certain symmetries have been used to simplify the problem. Reflection symmetry has been used to reduce the number of pairing configurations by a factor of 2. Charge inversion symmetry, where the valences of the carriers are inverted, reduces the problem by nearly another factor of 2 (some of the configurations map onto themselves under charge inversion followed by reflection).

A computer program was written in a symbolic manipulation language to perform the large number of relatively simple but recursively defined integrations required. The calculation of the contribution of a particular configuration is performed by finding the nonoverlapping volumes of the spheres of influence of each valence as a function of the position of the carriers, and integrating the exponentials of the volumes over the allowed positions of the carriers. The integration region for each configuration is bounded by the requirements that the carriers indeed pair as the configuration dictates, that at least one carrier of each carrier pair except the central one be inside a sphere of influence of another carrier pair, and that the carriers recombine in such an order that the central carrier pair recombines last. For this one-dimensional problem, these requirements can be expressed as a set of linear inequalities bounding the region of integration. These inequalities were incorporated into the integrand by converting them into Heaviside step functions

$$H(x)=\begin{cases}0, & x<0\\ 1, & x>0\end{cases}. \tag{23}$$

In addition to the step functions expressing the limits of integration, the expressions for the nonoverlapping volumes of the spheres of influence also contain step functions. The integral was then performed using the relation (assuming $a<b$)

TABLE II. Summary of NAN isolated clusters: three-pair configurations.

Pairs	Configuration	Notation	Density (ppm)
3		$P_{3\text{-}1}$	1620
3		$P_{3\text{-}2}$	1424
3		$P_{3\text{-}3}$	366
3		$P_{3\text{-}4}$	1880
3		$P_{3\text{-}5}$	206
3		$P_{3\text{-}6}$	1953
3		$P_{3\text{-}7}$	651
3		$P_{3\text{-}8}$	10290
3		$P_{3\text{-}9}$	620
3		$P_{3\text{-}10}$	12442
3		$P_{3\text{-}11}$	1158
3		$P_{3\text{-}12}$	24
3		$P_{3\text{-}13}$	3430
3		$P_{3\text{-}14}$	2396
3		$P_{3\text{-}15}$	4148
3		$P_{3\text{-}16}$	5766
3		$P_{3\text{-}17}$	1864
3		$P_{3\text{-}18}$	386
3		$P_{3\text{-}19}$	3412
3		$P_{3\text{-}20}$	3472
3		$P_{3\text{-}21}$	72
3		$P_{3\text{-}22}$	1302

$$\int_a^b H(c_0+c_1x_1+c_2x_2+c_3x_3+\cdots)g\,dx_1=\begin{cases}H(c_0+c_2x_2+c_3x_3+\cdots)\int_a^b g\,dx_1 & \text{if } c_1=0,\\ H(a-x_0)\int_a^b g\,dx_1+H(x_0-a)H(b-x_0)\int_{x_0}^b g\,dx_1 & \text{if } c_1\neq 0,\end{cases} \tag{24}$$

where x_0 is the root of the equation $c_0+c_1x_0+c_2x_2+c_3x_3+\cdots=0$, and g can be a function of all of the x's. Because this procedure nearly doubles the number of integrals for each inequality so resolved, a powerful automated procedure for performing the integrals was developed.

This integration procedure was applied to all possible configurations involving two- and three-pair clusters. The two-pair cluster in one dimension gives the result

$$P_{1,2}(x)=(\tfrac{4}{3}x^2+4x-3)e^{-4x}+\tfrac{9}{5}e^{-14x/3}+e^{-6x}+\tfrac{1}{5}e^{-8x}. \tag{25}$$

The three-pair cluster in one dimension gives

$$P_{1,3}(x)=(\tfrac{4}{15}x^4+\tfrac{8}{3}x^3+2x^2-6x+\tfrac{10}{63})e^{-4x}+(\tfrac{22}{15}x^2+\tfrac{168}{25}x-\tfrac{7623}{1000})e^{-14x/3}+\tfrac{625}{216}e^{-24x/5}+\tfrac{729}{272}e^{-44x/9}$$
$$+\tfrac{512}{225}e^{-5x}-\tfrac{3125}{792}e^{-26x/5}+\tfrac{1917}{1400}e^{-16x/3}+(\tfrac{4}{3}x^2+\tfrac{10}{3}x+\tfrac{34}{27})e^{-6x}+\tfrac{3033}{4400}e^{-20x/3}$$
$$+(\tfrac{2}{5}x^2+\tfrac{124}{225}x-\tfrac{1}{500})e^{-8x}+\tfrac{1881}{11\,900}e^{-26x/3}+\tfrac{83}{1050}e^{-10x}+\tfrac{7}{900}e^{-12x}\ . \tag{26}$$

When these expressions are combined with the result for $P_{1,1}(x)$, the power-series expansion for $P_{\mathrm{NAN},1}$ becomes, exact to fifth order in x,

$$P_{\mathrm{NAN},1}(x)=2-8x+\tfrac{64}{3}x^2-\tfrac{416}{9}x^3+\tfrac{3952}{45}x^4$$
$$-\tfrac{102\,592}{675}x^5+\cdots\ . \tag{27}$$

This is equivalent to

$$Q_{\mathrm{NAN},1}(x)=1-2x+4x^2-\tfrac{64}{9}x^3+\tfrac{104}{9}x^4$$
$$-\tfrac{3952}{225}x^5+\tfrac{51\,296}{2025}x^6-\cdots\ . \tag{28}$$

The carriers contained in clusters of up to three carrier pairs correspond to about 68% of the total carrier density. These carriers have an average recombination distance of about 40% of the inverse density, and therefore reflect the behavior of the early stages of recombination. A more detailed analysis of the calculations performed to arrive at these results shows that with only a little more work, one may account for a larger fraction of the carriers (74%). This method, however, does not give more terms in the expansion for $Q_{\mathrm{NAN},1}(x)$, and will not be presented here.

Including the two- and three-pair clusters is important in the NAN calculation, because the one-pair cluster alone is not indicative of NAN behavior. Indeed, the zeroth-order term in the expansion of $Q_{\mathrm{NAN},1}(x)$, the x^0 term, shows only the normalization of the distribution. The first-order term gives the contribution of the neighbors, or the linear density effect. The second-order term reflects the nearest-neighbor nature of the NAN scheme. It is only in the third-order term in $Q_{\mathrm{NAN},1}(x)$, the x^3 term, that the NAN competition starts to take effect. This first truly NAN term disagrees with both of the Dunstan ansatz solutions.

The higher order terms disagree progressively more with the Dunstan ansatz for $Q(x)$. The result of this disagreement is that the distributions are similar only for small x, and diverge increasingly for larger x. This behavior is exactly that seen in previous Monte Carlo results.[6]

In two and three dimensions, the expansion method fails to provide exact results because the integrals are intractable, and ordering schemes such as used for the one-dimensional case do not apply. One may recover some information, however, by calculating only the first term in the expansion for the two-pair clusters. In this case, the exponentials given in Eq. (20) may be replaced by their zeroth-order expansion, namely unity. This simplifies the calculation to the derivation of the volume of the $2n$-dimensional volume available for the two-pair NAN clusters. With this method, the three-dimensional calculation of the first truly NAN term can be found exactly, while the two-dimensional case reduces to a single integral which must be done numerically. The power series resulting from this calculation are

$$Q_{\mathrm{NAN},2}(x)=1-\pi x^2+\pi^2x^4-(0.409\,936\,74)\pi^3x^6+\cdots\ ,$$
$$Q_{\mathrm{NAN},3}(x)=1-\tfrac{4}{3}\pi x^3+\tfrac{16}{9}\pi^2x^6-\tfrac{7808}{8505}\pi^3x^9+\cdots\ . \tag{29}$$

Both of these series disagree with previous analytic approximations to the NAN distribution.[2,5]

III. PROPOSED EXACT SUM

Because so many terms in the expansion of $Q_{\mathrm{NAN},1}$ are available, it is tempting to look for a pattern and a sum of the series. We assert that we have found a simple pattern which can be summed exactly, and which provides an asymptotic solution predicted by previous Monte Carlo calculations. If one takes the logarithmic derivative of the partial power series given in Eq. (28), the result is

$$\frac{d\ln Q}{dx}=-2+4x-\tfrac{16}{3}x^2+\tfrac{16}{3}x^3-\tfrac{64}{15}x^4+\tfrac{128}{45}x^5-\cdots\ . \tag{30}$$

This series matches the simple pattern, which, when extrapolated to infinite terms, gives

$$\frac{d\ln Q}{dx}=\sum_{k=0}^{\infty}\frac{(-1)^{k+1}2^{2k+1}}{(k+1)!}x^k=\frac{1}{2x}(e^{-4x}-1)$$
$$=-2I_0(4x)\ . \tag{31}$$

Solution of the resulting differential equation gives

$$Q(x)=\exp[-2xI_1(4x)]\ . \tag{32}$$

This relation is shown in Fig. 2 as a solid curve. This expression has by construction the correct power series expansion to the number of terms given in Eq. (28), and has the asymptotic dependence

$$Q(x)\sim\tfrac{1}{2}e^{-\gamma/2}x^{-1/2}\ . \tag{33}$$

The power-law asymptotic dependence matches exactly the result of Monte Carlo calculations in both exponent and prefactor, indicating that the proposed power series may in fact be the correct one. Indeed, stringent lower bounds to higher-order terms in the power series for $Q_{\mathrm{NAN},1}$, produced by partial sums of the four-pair clusters, correspond nearly exactly to the higher terms in the expansion of Eq. (32). It is interesting to compare the differential equation implied by this method to the one used by Dunstan. A simple attempt to generalize this

solution to higher dimensions, by replacing $2x \rightarrow v_n x^n$, following the hypothesis of Searle and Bishop for the asymptotic dependence, fails here to reproduce the known low-order terms in the NAN power series given in Eq. (29).

IV. SUMMARY

The recombination distance distribution in the distant-pair model has been investigated. The distant-pair model has proven to be a remarkably difficult model to solve analytically. The solutions given in the literature are shown to be based on inaccurate approximations. An analytic method for the derivation of the power series for the nearest-available-neighbor distribution has proven successful in the one-dimensional case, and has provided some results in two and three dimensions. A proposed exact sum of the one-dimensional power series gives excellent agreement with previous Monte Carlo results. All of the results demonstrate that previous analytic methods underestimate the mean recombination distance and hence the number of metastable carriers in the distant-pair model.

ACKNOWLEDGMENTS

We thank R. Barakat for his helpful comments, and acknowledge support from the U.S. Joint Services Electronics Program under Contract No. N00014-84-K-0465 and the U.S. Department of Energy (DOE) under DOE Basic Sciences Program Contract No. DE-FG02-84ER45070. One of us (J.R.E.) acknowledges support from I.B.M.

APPENDIX: DECAY FUNCTIONS

1. The decay function $I_n(\theta)$

The decay function $I_n(\theta)$ introduced in the calculation of the distant-pair decay is given as

$$I_n(\theta)=\int_0^\infty z^n e^{-z}\exp(-\theta e^{-z})dz \ . \tag{A1}$$

Onsager and Stewart[10] have given some properties of $K(\theta)=-\frac{1}{3}\theta I_3(\theta)$. In this appendix, we present the generalization of their results to arbitrary dimension n, and give some further properties of $I_n(\theta)$ and of the incomplete decay integral $I_n(\theta,z)$.

The differential and integral properties of $I_n(\theta)$ are easily deduced from its definition using integration by parts.

$$\frac{d}{d\theta}I_n(\theta)=\theta^{-1}[nI_{n-1}(\theta)-I_n(\theta)] \quad (n>0) \ , \tag{A2}$$

$$\int_0^\theta I_n(\theta')d\theta'=\theta I_{n+1}(\theta)/(n+1) \ . \tag{A3}$$

By expanding the exponential in the integrand in Eq. (A1), an infinite series for $I_n(\theta)$ valid for small θ may be written:

$$I_n(\theta)=n!\sum_{k=1}^{\infty}\frac{(-\theta)^{k-1}}{k^n k!} \ . \tag{A4}$$

For $n=0$ this simplifies to

$$I_0(\theta)=\frac{1}{\theta}(1-e^{-\theta}) \ , \tag{A5}$$

and for $n=1$ it becomes

$$I_1(\theta)=\frac{1}{\theta}[\ln\theta+\gamma+E_1(\theta)] \ , \tag{A6}$$

where $\gamma=0.577\ldots$ is Euler's constant and $E_1(\theta)$ is the exponential integral of the first order.

For large values of θ, the series in Eq. (A4) converges rather slowly. Following Onsager and Stewart, one may find an expression suitable for large θ by making the substitution $u=\theta e^{-z}$ in Eq. (A1). Then

$$I_n(\theta)=M_n(\theta)+N_n(\theta) \ , \tag{A7}$$

where

$$M_n(\theta)=\frac{1}{\theta}\int_0^\infty \ln^n(\theta/u)e^{-u}du \tag{A8}$$

and

$$N_n(\theta)=-\frac{1}{\theta}\int_\theta^\infty \ln^n(\theta/u)e^{-u}du \ . \tag{A9}$$

$M_n(\theta)$ can eventually be written as

$$M_n(\theta)=\frac{n!}{\theta}\sum_{k=0}^{n}\frac{\Gamma_k}{(n-k)!}\ln^{n-k}\theta \ , \tag{A10}$$

where the coefficients Γ_k are given by

$$\Gamma(1-\mu)=\sum_{k=0}^{\infty}\Gamma_k\mu^k \tag{A11}$$

and can be calculated from the recursion relations[11]

$$\Gamma_0=1 \ , \tag{A12}$$

$$\Gamma_1=\gamma \ , \tag{A13}$$

$$\Gamma_{k+1}=\frac{1}{k+1}\sum_{m=0}^{k}s_{m+1}\Gamma_{k-m} \ , \tag{A14}$$

$$s_1=\gamma \ , \tag{A15}$$

$$s_m=\zeta(m) \ \text{ when } m>1 \ , \tag{A16}$$

where $\zeta(m)$ is the Riemann zeta function.

For $N_n(\theta)$ one obtains

$$N_n(\theta)=n!(-1)^{n+1}e^{-\theta}\sum_{m=0}^{\infty}\frac{b_{n,m}}{\theta(\theta+1)(\theta+2)\cdots(\theta+m)} \ , \tag{A17}$$

where the $b_{n,m}$ are all integers and are defined by

$$nb_{n,m}=\sum_{k=0}^{m}\binom{m}{k}b_{n-1,k}b_{1,m-k} \ , \tag{A18}$$

$$b_{1,m+1}=\sum_{k=0}^{m}a_{m,k} \ , \tag{A19}$$

$$b_{1,0}=0 \ , \tag{A20}$$

$$a_{m,k}=ma_{m-1,k}-ka_{m-1,k-1}\ , \tag{A21}$$

$$a_{0,0}=1\ , \tag{A22}$$

$$a_{m,k}=0 \quad \text{when } m<0 \text{ or } k<0\ . \tag{A23}$$

A table of the integers $b_{n,m}$ for $n=1$, 2, 3, and 4 and m, up to 15 is given in Table III.

The Laplace transform properties of $I_n(\theta)$ may be calculated simply. We have

$$\tilde{I}_n(\sigma)=\mathcal{L}\{I_n(\theta)\}=\int_0^\infty e^{-\sigma\theta}I_n(\theta)d\theta\ . \tag{A24}$$

Using the definition of $I_n(\theta)$ and performing the θ integration, this becomes

$$\tilde{I}_n(\sigma)=\int_0^\infty \frac{z^n}{\sigma e^z+1}dz=n!\sum_{k=1}^{\infty}(-1)^{k+1}\frac{1}{k^{n+1}\sigma^k}\ . \tag{A25}$$

The inverse Laplace transformation of $I_n(\theta)$ may be directly deduced from Eq. (A1) by using the substitution $\omega=e^{-z}$,

$$I_n(\theta)=\int_0^1 \ln^n\left[\frac{1}{\omega}\right]e^{-\theta\omega}d\omega\ , \tag{A26}$$

whereupon

$$\mathcal{L}^{-1}\{I_n(\theta)\}=\begin{cases}\ln^n(1/\omega) & \text{when } 0<\omega<1\\ 0 & \text{when } \omega>1\ ,\end{cases} \tag{A27}$$

by inspection.

We also require the generalized Laplace transform

$$\tilde{I}_{n,m}(\sigma)=\int_0^\infty e^{-\theta\sigma}\theta^m I_n(\theta)d\theta\ . \tag{A28}$$

This function has the differential property

$$\frac{d}{d\sigma}\tilde{I}_{n,m}(\sigma)=-\tilde{I}_{n,m+1}(\sigma)\ . \tag{A29}$$

After some integration, the expression for $\tilde{I}_{n,m}$ becomes

$$\tilde{I}_{n,m}(\sigma)=\frac{m!}{\sigma m}\int_0^\infty [nz^{n-1}+(m-1)z]\times e^{(m-1)z}(1+\sigma e^z)^{-m}dz\ , \tag{A30}$$

which gives the recurrence relation

$$\tilde{I}_{n,m}(\sigma)=\sigma^{-1}[n\tilde{I}_{n-1,m-1}(\sigma)+(m-1)\tilde{I}_{n,m-1}(\sigma)]\ . \tag{A31}$$

The recurrence terminates when $n=0$ or $m=0$. For these cases, we get

$$\tilde{I}_{n,0}(\sigma)=\tilde{I}_n(\sigma)\ , \tag{A32}$$

$$\tilde{I}_{0,m}(\sigma)=(m-1)![\sigma^{-m}-(1+\sigma)^{-m}]\ , \tag{A33}$$

$$\tilde{I}_{0,0}(\sigma)=\ln(1+\sigma)\ . \tag{A34}$$

2. The incomplete decay function $I_n(\theta,z)$

To calculate the recombination distance distribution in certain models, it is useful to define an incomplete decay function analogous to the incomplete gamma function. Let

$$I_n(\theta,z)=\int_z^\infty x^n e^{-x}\exp(-\theta e^{-x})dx \tag{A35}$$

be the incomplete decay function. Then

$$I_n(\theta,0)=I_n(\theta)\ . \tag{A36}$$

The differential and integral properties of $I_n(\theta,z)$ are

$$\frac{\partial}{\partial\theta}I_n(\theta,z)=\frac{1}{\theta}[nI_{n-1}(\theta,z)-I_n(\theta,z)]+z^n e^{-z}\exp(-\theta e^{-z})\ , \tag{A37}$$

$$\frac{\partial}{\partial z}I_n(\theta,z)=-z^n e^{-z}\exp(-\theta e^{-z})\ , \tag{A38}$$

TABLE III. Coefficients $b_{n,m}$ in the expansion of $N_n(\theta)$.

m \ n	1	2	3	4
1	1	0	0	0
2	0	1	0	0
3	1	0	1	0
4	1	4	0	1
5	8	5	10	0
6	26	58	15	20
7	194	217	238	35
8	1142	2035	1008	728
9	9736	13 470	11 611	3444
10	81384	134 164	85 410	47 815
11	823 392	1 243 770	983 059	387 090
12	8 738 016	14 129 410	10 021 671	5 099 204
13	104 336 880	164 244 808	127 015 018	56 743 687
14	1 328 270 880	2 151 576 620	1 607 649 134	797 684 160
15	18 419 317 968	29 671 566 836	23 003 592 532	10 934 343 875

$$\int_0^{\theta} I_n(\theta',z)d\theta' = \frac{\theta}{n+1} I_{n+1}(\theta,z) - \frac{z^{n+1}}{n+1}[1-\exp(-\theta e^{-z})] . \quad \text{(A39)}$$

For small θ one may expand the exponential in Eq. (A35) to get

$$I_n(\theta,z) = \sum_{k=1}^{\infty} \frac{(-\theta)^{k-1}}{k^n k!} \Gamma(n+1,kz) . \quad \text{(A40)}$$

Alternatively one may set $u = \theta e^{-x}$ in Eq. (A35) so that

$$I_n(\theta,z) = M_n(\theta) + e^{-z} \sum_{k=0}^{n} \begin{bmatrix} n \\ k \end{bmatrix} z^k N_{n-k}(\theta e^{-z}) , \quad \text{(A41)}$$

where $M_n(\theta)$ and $N_n(\theta)$ are defined as before. An equivalent expression could also be obtained from Eq. (A35) using the definition of the incomplete gamma function.

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