Donor fluorescence at high trap concentration. II.

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We continue our study of the time development of the donor fluorescence in the presence of a high concentration of acceptor ions which act as traps for the excitation. It is assumed that the transfer rates between donors are symmetric and independent of the energy mismatch between ions and that there is no back transfer from the traps. We study two classes of systems. In the first, the donor ions form a lattice with the traps distributed at random as interstitial impurities, each trap being coupled to a single donor. The effective decay rate is calculated in the coherent-potential approximation (CPA) and compared with a low-concentration theory based on the average t-matrix approximation (ATA). Results are presented for one, two, and three dimensions. In three dimensions the ATA and CPA are in close agreement up to a trap concentration $c_A \approx 0.01$. Beyond that, the decay rate calculated in the CPA increases more rapidly with increasing trap concentration than the corresponding rate in the ATA. In one and two dimensions the asymptotic fluorescence decays more rapidly than the algebraic decay calculated in the ATA. At low concentration the effective decay rate varies as c_A^2 in one dimension and $c_A/|\ln c_A|$ in two dimensions. The behavior in the diffusion regime is studied in a system where the donor-acceptor transfer varies as r^{-6}. A self-consistent equation for the t matrix is solved numerically, and results are obtained for the variation of the decay rate and effective diffusion constant with trap concentration.

I. INTRODUCTION

The time development of optical fluorescence in the presence of a random distribution of traps has been the subject of a recent series of theoretical papers.[1–4] In Ref. 1 a general theory of the fluorescence was developed which made use of the average t-matrix approximation (ATA) to calculate the Laplace transform of the integrated donor fluorescence following broad-band excitation. In Ref. 2 the theory was applied to the calculation of the fluorescence in a system where the fluorescing ions (donors) formed a regular array with the traps (acceptors) being substitutional impurities. Nearest-neighbor incoherent transfer between donors and from donors to acceptors was assumed. The opposite limit, where there is a large number of donors in the sphere of influence of an acceptor, was considered in Ref. 3. In this regime a continuum description based on the diffusion equation is appropriate.

The analysis developed in Refs. 1–3 is applicable to systems where the concentration of traps is much less than the donor concentration. In order to determine the limits of validity of the low-concentration theory it is necessary to study the donor fluorescence as a function of trap concentration. In Ref. 4 an analysis of the donor fluorescence was carried out for arbitrary values of the ratio of acceptor to donor concentration. It was assumed that the donor and acceptor arrays were dilute and that both the donor-donor and donor-acceptor transfer rates varied as r^{-6} characteristic of dipole-dipole transfer. The coherent-potential approximation (CPA) was utilized to calculate the effective decay rate as a function of the ratio $n_A\alpha^{1/2}/(n_D\beta^{1/2})$. Here n_A and n_D denote the acceptor and donor concentrations, respectively, while α and β are the coefficients multiplying r^{-6} in the corresponding donor-acceptor and donor-donor transfer rates. According to the theory, which is applicable only when $\alpha \lesssim \beta$, the ATA is reasonably accurate for concentrations such that $n_A\alpha^{1/2}/(n_D\beta^{1/2}) \lesssim 0.2$. Beyond that point the asymptotic decay rate begins to depart significantly from its linear dependence on trap concentration.

This paper is a continuation of the analysis which was begun in Ref. 4. We investigate the dependence of the donor fluorescence on trap concentration in a system where the donor ions form a lattice. The traps are "interstitial" impurities which have the property that each trap can interact with a single donor and that a donor ion can transfer to no more than one trap. The donor fluorescence is calculated using the CPA and comparison is made with the results obtained with the ATA. Although the model is somewhat artificial we have reason to believe that the qualitative features of many of our results apply also to more realistic situations such as considered in Ref. 2.

The CPA can be understood as an approximation in which trapping is characterized by a self-consistent,

frequency-dependent transfer rate. The self-consistent equation amounts to requiring that the single-trap t matrix associated with the difference between the actual and the effective transfer rates vanishes on the average [cf. Eq. (2.7)]. As will be shown below the CPA reproduces the exact results for the reciprocal of the Laplace transform of the time-dependent fluorescent decay at high and low trap concentrations. In addition, it provides a reasonable interpolation formula between these limits.

In addition to our work on short-range interactions we have studied the effects of a finite concentration of traps in the diffusion regime considered in Ref. 3. We propose a self-consistent integral equation for the t matrix which we solve using numerical techniques developed in Ref. 3. Specializing to the case where the donor-acceptor transfer rate is given by αr^{-6} we calculate the asymptotic decay rate and the effective diffusion constant as a function of the dimensionless parameter $n_A(\alpha/D)^{3/4}$, D denoting the diffusion constant.

The analysis of systems with interstitial traps is outlined in Sec. II. In Sec. III we discuss the diffusion model and in Sec. IV we comment on our results.

II. INTERSTITIAL TRAPS

In this section we analyze the effect of a finite concentration of traps on the integrated fluorescence following broad-band excitation of the donors. We assume that the donors form a lattice. A fraction c_A have interstitial traps associated with them. Each trap can receive excitation from its companion donor; there is no backtransfer from the trap to the donor. In the interest of simplicity we assume that the donor-donor transfer takes place only between nearest neighbors. The transfer is incoherent, symmetric, and independent of the energy mismatch between donors.

We follow the notation of Ref. 1 and write the normalized intensity of the donor fluorescence $F(t)$ as

$$F(t)=e^{-\gamma_R t}f(t) \ , \tag{2.1}$$

where γ_R^{-1} is the lifetime of the fluorescing level. The function $f(t)$ characterizes the loss in intensity due to one-way transfer to traps.

When $c_A \ll 1$ the ATA can be used to calculate the Laplace transform of $f(t)$ which we define by

$$\hat{f}(s)=\int_0^\infty dt\, e^{-st}f(t) \ . \tag{2.2}$$

We obtain the result[1]

$$\hat{f}(s)=[s+c_A X/[1+Xg_0(s)]]^{-1} \ . \tag{2.3}$$

Here X denotes the donor-acceptor transfer rate which is assumed to be the same for all donor-acceptor pairs. The function $g_0(s)$ is given by the expression

$$g_0(s)=\frac{1}{N}\sum_{\vec{k}}\frac{1}{s+zW(1-\gamma_k)} \ , \tag{2.4}$$

in which

$$\gamma_k=z^{-1}\sum_j{}'\exp(i\vec{k}\cdot\vec{r}_{ij}) \ , \tag{2.5}$$

where $\sum_j'$ denotes a sum over the z nearest neighbors of the site i, and W is the donor-donor transfer rate. The symbol N is the number of donors, and the sum on $\vec{k}$ is over the Brillouin zone associated with the donor lattice.

The application of the CPA to the interstitial problem is similar to the application made in Ref. 4. The function $\hat{f}(s)$ is given by

$$\hat{f}(s)=[s+X_{\rm CPA}(s)]^{-1} \ , \tag{2.6}$$

where $X_{\rm CPA}(s)$ is the solution to the equation

$$\int\frac{d\hat{X}\,\mathcal{P}(\hat{X})[\hat{X}-X_{\rm CPA}(s)]}{1+[\hat{X}-X_{\rm CPA}(s)]G_0(s)}=0 \ . \tag{2.7}$$

Here $\mathcal{P}(\hat{X})$ is the probability distribution for the random variable $\hat{X}$ characterizing the donor-acceptor transfer. For the model under consideration we have

$$\mathcal{P}(\hat{X})=c_A\delta(\hat{X}-X)+(1-c_A)\delta(\hat{X}) \ , \tag{2.8}$$

assuming a fraction c_A of the donors can transfer to traps at the rate X. The Green's function in (2.7) is given by an expression similar to (2.4), i.e.,[5]

$$G_0(s)=\frac{1}{N}\sum_{\vec{k}}\frac{1}{s+X_{\rm CPA}(s)+zW(1-\gamma_k)} \ . \tag{2.9}$$

With the distribution (2.8) Eq. (2.7) reduces to

$$X_{\rm CPA}=\tfrac{1}{2}\{Q+X-[(Q+X)^2-4c_AXQ]^{1/2}\} \ , \tag{2.10}$$

with

$$Q(s)=G_0(s)^{-1} \ . \tag{2.11}$$

In the limit $c_A\to 0$ Eq. (2.10) reduces to the ATA result shown in Eq. (2.3),

$$X_{\rm CPA}(s)\to\frac{c_AX}{1+XG_0(s)}\to\frac{c_AX}{1+Xg_0(s)} \qquad (c_A\ll 1,\ s \text{ finite}) \ , \tag{2.12}$$

since in this limit $G_0(s)=g_0(s)$. In the opposite limit, $c_A=1$, we have

$$X_{\rm CPA}(s)=X \quad (c_A=1) \ . \tag{2.13}$$

The result shown in Eq. (2.13) is to be expected since when $c_A=1$ all donors are associated with traps. Under these conditions donor-donor transfer has no influence on the effective rate of transfer to the traps.

We have solved Eq. (2.10) in the limit $s=0$ for one-, two-, and three-dimensional lattices.[6] In all cases we obtain a finite value for $X_{\mathrm{CPA}}(0)$, which we identify with an effective decay rate. In Fig. 1 we display our results for $X_{\mathrm{CPA}}(0)$ for a simple cubic lattice along with the predictions of the ATA for the cases $X=W$ and $X=1000W$. It is apparent that the decay rates calculated in the ATA and CPA are close to one another for $c_A \lesssim 0.01$. The curves diverge with increasing trap concentration with the larger discrepancy occurring for $X=1000W$.

In Refs. 1 and 2 it was pointed out that in one and two dimensions $g_0(s)$ diverged as $s^{-1/2}$ and $-\ln s$, respectively, in the $s\to 0$ limit. As a consequence of this divergence $f(t)$ in the ATA has the asymptotic behavior

$$f(t) \sim (4\pi c_A^2 Wt)^{-1/2} \tag{2.14}$$

in one dimension, and

$$f(t) \sim (4\pi c_A Wt)^{-1} \tag{2.15}$$

in two dimensions.

As noted, in the CPA $X_{\mathrm{CPA}}(s)$ remains finite in the limit $s\to 0$. The singular behavior of the Green's function which gives rise to the algebraic decay of $f(t)$ leads in the CPA to effective decay rates which are nonlinear functions of the concentration in the limit $c_A \to 0$. To see how this comes about we note that (2.10) becomes

$$X_{\mathrm{CPA}}(0) = \frac{c_A X}{1+XG_0(0)} \ , \tag{2.16}$$

for $s=0$, $c_A \ll 1$. In one dimension $G_0(0)$ is given by

$$\begin{aligned} G_0(0) &= \frac{1}{\pi}\int_0^{\pi} \frac{dy}{X_{\mathrm{CPA}}(0)+2W(1-\cos y)} \\ &= \{[X_{\mathrm{CPA}}(0)+2W]^2-4W^2\}^{-1/2} \\ &\approx [4X_{\mathrm{CPA}}(0)W]^{-1/2} \ . \end{aligned} \tag{2.17}$$

Combining (2.16) and (2.17) we obtain the result

$$X_{\mathrm{CPA}}(0) = 4c_A^2 W \ , \tag{2.18}$$

which holds when $c_A \ll 1, X/16W$. From this equation it is evident that the effective decay rate is independent of X and varies as the square of the concentration. More generally it can be shown that in the same limit $f(t)$ is a universal function of the dimensionless variable $c_A^2 Wt$. In Fig. 2 we show the variation of $X_{\mathrm{CPA}}(0)$ with trap concentration over the entire range $0 \leq c_A \leq 1$.

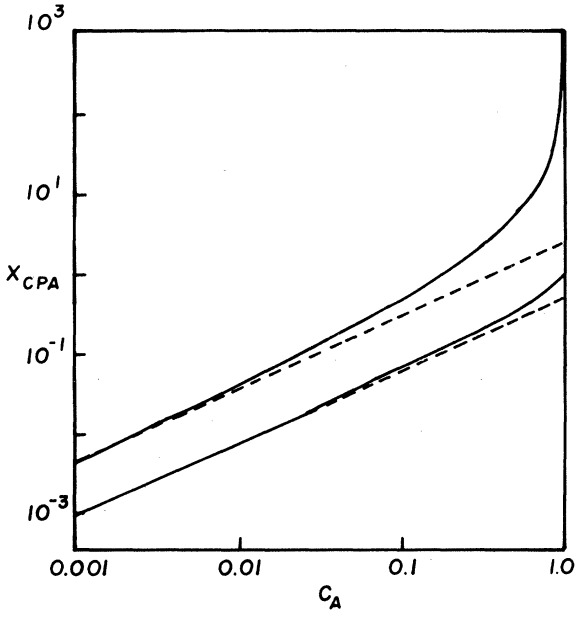

FIG. 1. Effective decay rate $X_{\mathrm{CPA}}(0)$ for a simple cubic lattice as a function of trap concentration c_A. $X_{\mathrm{CPA}}(0)$ is measured in units of the donor-donor transfer rate W. The upper solid line is for $X=1000W$; the lower solid line is for $X=W$. In both cases the broken line is the result given by the ATA.

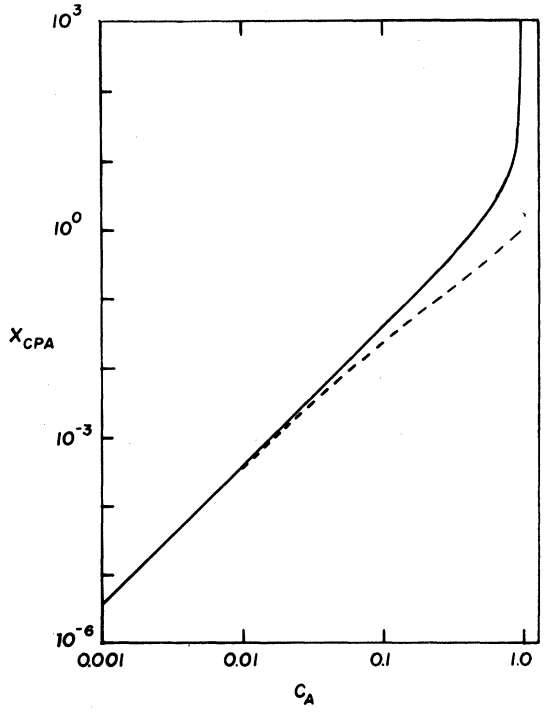

FIG. 2. Effective decay rate $X_{\mathrm{CPA}}(0)$ for a one-dimensional lattice as a function of trap concentration c_A. $X_{\mathrm{CPA}}(0)$ is measured in units of the donor-donor transfer rate W. The solid line is for $X=1000W$; the broken line is for $X=W$.

We can obtain an approximate expression for $f(t)$ at long times by expanding $X_{\mathrm{CPA}}(s)$ in powers of s. Keeping only the zeroth and first-order terms we obtain

$$f(s) \approx [s + X_{\mathrm{CPA}}(0) + X'_{\mathrm{CPA}}(0)s]^{-1} , \quad (2.19)$$

where $X'_{\mathrm{CPA}}(0) = dX_{\mathrm{CPA}}(0)/ds$. From (2.19) we obtain

$$f(t) \sim [1 + X'_{\mathrm{CPA}}(0)]^{-1} \exp\left[-\frac{X_{\mathrm{CPA}}(0)t}{1 + X'_{\mathrm{CPA}}(0)}\right] . \quad (2.20)$$

At low concentration in one dimension $X'_{\mathrm{CPA}}(0) \approx 1$, whereas in all dimensions $X'_{\mathrm{CPA}}(0) = 0$ when $c_A = 1$ [cf. Eq. (2.13)].

In the case of the square lattice we have

$$G_0(0) = \frac{1}{\pi^2}\int_0^{\pi} dx \int_0^{\pi} dy\, [X_{\mathrm{CPA}}(0) + 4W(1 - \tfrac{1}{2}\cos x - \tfrac{1}{2}\cos y)]^{-1} = (\pi a W)^{-1} F(4/a^2) , \quad (2.21)$$

where

$$a = 2 + X_{\mathrm{CPA}}(0)/2W , \quad (2.22)$$

and $F(m)$ is the complete elliptic integral of the first kind defined by

$$F(m) = \int_0^{\pi/2} (1 - m\sin^2\theta)^{-1/2}\, d\theta . \quad (2.23)$$

When $X_{\mathrm{CPA}}(0) \ll W$, $G_0(0)$ reduces to

$$G_0(0) = (4\pi W)^{-1} \ln[32W/X_{\mathrm{CPA}}(0)] . \quad (2.24)$$

Combining this with (2.16) we obtain the result

$$X_{\mathrm{CPA}}(0) = \frac{4\pi c_A W}{\ln[32W/X_{\mathrm{CPA}}(0)]} , \quad (2.25)$$

from which we infer that $X_{\mathrm{CPA}}(0)$ varies as $c_A/|\ln c_A|$ in the limit $c_A \to 0$. Similarly it can be shown that $X'_{\mathrm{CPA}}(0)$ vanishes as $[\ln(32W/X_{\mathrm{CPA}}(0)]^{-1}$ in the same limit. Our results for $X_{\mathrm{CPA}}(0)$ in two dimensions are displayed in Fig. 3.

It must be emphasized that Eqs. (2.18) and (2.25) hold only for $s = 0$ and thus characterize the integral $\int_0^{\infty} f(t)\, dt$. In view of the comments made earlier about the close connection between the ATA and the CPA at low trap concentrations we expect that for large s or equivalently for short times the ATA and the CPA will give similar results when $c_A \ll 1$. The two approximations will begin to differ when $G_0(s)$ departs from $g_0(s)$. Very crudely, this will happen when

$$s \approx X_{\mathrm{CPA}}(0) , \quad (2.26)$$

or at a time $t \sim X_{\mathrm{CPA}}(0)^{-1}$ (see Fig. 4). Thus when $c_A \ll 1$ $f(t)$ will be given equally well by either the ATA or the CPA for $0 \ll t \ll X_{\mathrm{CPA}}(0)^{-1}$ whereas for $t \gg X_{\mathrm{CPA}}(0)^{-1}$ the CPA is the superior approximation.

Recently, Movaghar and Sauer[7] succeeded in obtaining an exact solution for $\hat{f}(s)$ when $X/W = \infty$ for a one-dimensional array with an arbitrary concentration of traps. Inverting the transform they compare the CPA expression for $f(t)$ with the exact result for $c_A = 0.01$, 0.1, and 0.6. Over the interval $0 \leq c_A^2 Wt \lesssim 1$ they find that the CPA overestimates the rate of decay of the fluorescence. However for $c_A \ll 1$ and $c_A^2 Wt \gg 1$ the CPA is significantly closer to the exact result than is the corresponding expression calculated in the ATA, where $f(t)$ decays much less rapidly. This is evident from a comparison of values for the integral $\int_0^{\infty} f(t)\, dt$ which for the exact $f(t)$ is equal to $(2c_A^2 W)^{-1}$ when $X/W = \infty$. In the ATA the integral is infinite, whereas in the CPA it has the value $(1 + c_A)/(4c_A^2 W)$. Although the CPA is superior to the ATA the first passage time result $f(t) = \exp[-4c_A(Wt/\pi)^{1/2}]$, is actually a better approximation than the CPA when $c_A < 0.5$. For $c_A > 0.5$ the situation is reversed, with the CPA being the preferred approximation.

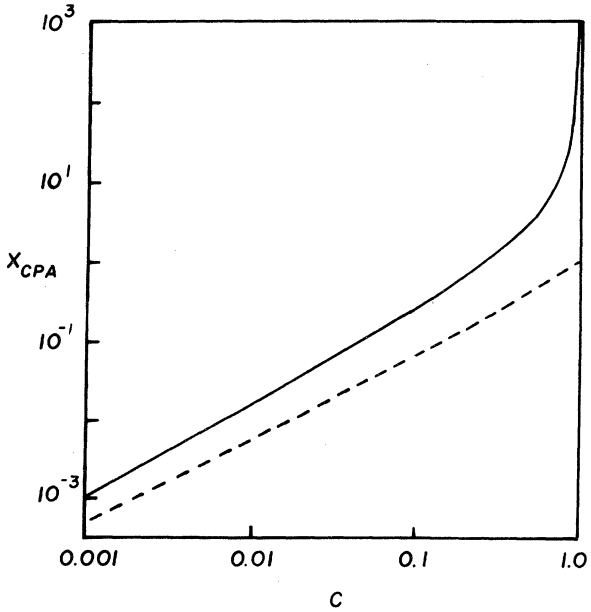

FIG. 3. Effective decay rate $X_{\mathrm{CPA}}(0)$ for a square lattice as a function of trap concentration c_A. $X_{\mathrm{CPA}}(0)$ is measured in units of the donor-donor transfer rate W. The solid line is for $X = 1000W$; the broken line is for $X = W$.

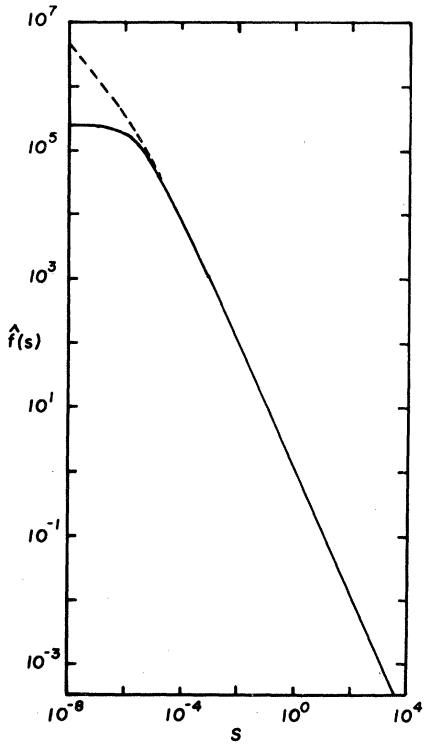

FIG. 4. Log-log plot of $\hat{f}(s)$ vs s for a one-dimensional lattice with $c_A = 0.001$ and $X = 1000W$. The variable s is measured in units of W and $\hat{f}(s)$ in units of W^{-1}. The solid line is the CPA result; the broken line is obtained using the ATA. The two curves begin to deviate at the point where $s \approx X_{\mathrm{CPA}}(0) = 4 \times 10^{-6}$.

III. DIFFUSION LIMIT

As discussed in Ref. 1 the diffusion approach is appropriate whenever there is a large number of donors in the sphere of influence of an acceptor. In systems where the donor-acceptor transfer rate is αr^{-6} and the donor-donor rate is βr^{-6} the diffusion regime corresponds to $\alpha >> \beta$. The time dependence of the fluorescence in this regime has been treated within the framework of the ATA in Ref. 3. In Refs. 1 and 3 it is shown that in the diffusion regime $\hat{f}(s)$ can be written

$$\hat{f}(s) = \left(+ n_A \int d\vec{r}\; T(\vec{r},s) \right)^{-1} , \tag{3.1}$$

where n_A is the acceptor concentration. In the ATA the function $T(\vec{r},s)$ is a solution to the integral equation

$$T(\vec{r},s) = V(r) - \int d\vec{r}\,'\, V(r) g_0(\vec{r}-\vec{r}\,',s) T(\vec{r}\,',s) \;; \tag{3.2}$$

in which $V(r)$ is the donor-acceptor transfer rate.

The function $g_0(\vec{r}-\vec{r}\,',s)$ is given by

$$g_0(\vec{r}-\vec{r}\,',s) = \frac{1}{(2\pi)^3} \int \frac{d\vec{k} \exp[i\vec{k}\cdot(\vec{r}-\vec{r}\,')]}{s + Dk^2} , \tag{3.3}$$

where the integral is over all of $\vec{k}$ space and D denotes the donor diffusion constant.

Equations (3.1)–(3.3) apply in the low-concentration regime which in the case where $V(r) = \alpha r^{-6}$ corresponds to $n_A(\alpha/D)^{3/4} << 1$. In order to study the behavior at higher concentrations we make a modification of (3.3) which is suggested by the analysis in the Appendix to Ref. 1.

$$s + Dk^2 \rightarrow s + Dk^2 + n_A \int d\vec{r}\; e^{i\vec{k}\cdot\vec{r}} T(\vec{r},s) , \tag{3.4}$$

which to order k^2 is equivalent to

$$s + Dk^2 \rightarrow s + \gamma(s) + D_{\mathrm{eff}}(s) k^2 , \tag{3.5}$$

where $\gamma(s)$ and $D_{\mathrm{eff}}(s)$, given by

$$\gamma(s) = n_A \int d\vec{r}\; T(\vec{r},s) , \tag{3.6}$$

and

$$D_{\mathrm{eff}}(s) = D - \tfrac{1}{6} n_A \int d\vec{r}\; r^2 T(\vec{r},s) , \tag{3.7}$$

are the s-dependent decay rate and effective diffusion constant, respectively.

This approach has a simple physical interpretation. The function $g_0(\vec{r}-\vec{r}\,',s)$ in (3.2) is the propagator for the donor excitation. Our approximation amounts to replacing the bare propagator by an effective propagator characterizing the transfer of excitation in a lossy medium in which the presence of the traps also influences the rate of diffusion. Equation (3.2) with g_0 replaced by

$$g_M(\vec{r}-\vec{r}\,',s) = \frac{1}{(2\pi)^3} \int \frac{d\vec{k} \exp[i\vec{k}\cdot(\vec{r}-\vec{r}\,')]}{s + \gamma(s) + D_{\mathrm{eff}}(s) k^2} , \tag{3.8}$$

then becomes a self-consistent equation for $T(\vec{r},s)$.

We have solved this equation iteratively in the limit $s = 0$ for the transfer rate $V(r) = \alpha r^{-6}$ $(r_c \leq r \leq \infty)$, $V(r) = 0$ $(r < r_c)$ using numerical techniques discussed in Ref. 3. The calculations were carried out with values of $\alpha(5.4 \times 10^{-38}\ \mathrm{cm^6\,s^{-1}})$, $D(14.0 \times 10^{9}\ \mathrm{cm^2\,s^{-1}})$, and $r_c(3.1 \times 10^{-8}\ \mathrm{cm})$ appropriate to the $^3P_0$ fluorescence of Pr^{3+} at 12.5 K in $Pr_{0.95}Nd_{0.05}F_3$.[3]

Our results for $\gamma(0)$ are plotted in Fig. 5 as a function of $\tilde{c}_A \equiv n_A(\alpha/D)^{3/4}$. The broken line is the prediction of the ATA which is seen to be in reasonable

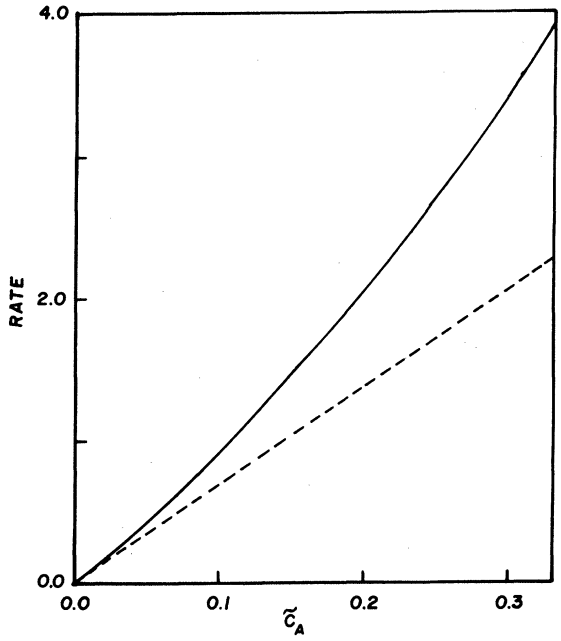

FIG. 5. Asymptotic decay rate $\gamma(0)$ in the diffusion limit as a function of $\tilde{c}_A = n_A(\alpha/D)^{3/4}$. The broken line shows the predictions of the linear theory. The decay rates are measured in units of $(D^3/\alpha)^{1/2}$.

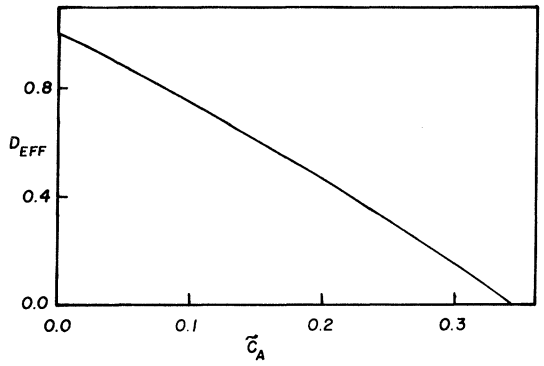

FIG. 6. Effective diffusion constant $D_{\text{eff}}(0)$ as a function of $\tilde{c}_A = n_A(\alpha/D)^{3/4}$. D_{eff} is measured in units of D the donor diffusion constant for $\tilde{c}_A = 0$.

agreement with the CPA out to $\tilde{c}_A \approx 0.1$. The corresponding behavior of $D_{\text{eff}}(0)$ is shown in Fig. 6 where we have plotted $D_{\text{eff}}(0)$ vs $\tilde{c}_A$. The effective diffusion constant decreases nearly linearly with concentration dropping to zero at $\tilde{c}_A \approx 0.33$ at which point the theory loses all meaning. The breakdown of the theory with increasing trap concentration is not surprising since our modified t-matrix equation is the counterpart of the low-concentration approximation to Eq. (2.10), $X_{\text{CPA}}(s) = c_A X/[1 + XG_0(s)]$.

IV. DISCUSSION

The results presented in Ref. 4 and in Secs. II and III of this paper show that the asymptotic decay of the donor fluorescence at high trap concentration is more rapid than is predicted by the linear theory (ATA). This is especially noticeable in one- and two-dimensional systems where algebraic decay is predicted by the ATA. In the case of a simple cubic lattice we found that the linear theory works well for $c_A \leq 0.01$. At higher concentrations there was a noticeable deviation between the CPA and ATA decay rates with the greater discrepancy occurring for the larger value of X/W. Figure 5 shows that in the diffusion regime the ATA works well for $\tilde{c}_A \lesssim 0.1$. As a point of reference we note that the value of $\tilde{c}_A$ for the $^3P_0$ fluorescence in $Pr_{0.95}Nd_{0.05}F_3$ at 12.5 K is 0.03.

Although there has been comparatively little systematic experimental work on the effect of trap concentration on donor fluorescence we believe that such studies are worthwhile. Measurements of the asymptotic decay rates for different numbers of traps should reveal the nonlinear behavior discussed in this paper. The one-dimensional systems at low trap concentration are particularly interesting.

In connection with the theory it should be emphasized that the CPA has proved to be a highly successful approach to problems like the one under consideration here which involve diagonal disorder.[8] For this reason an extension of the theory to cover substitutional impurities of the type discussed in Ref. 2 would be especially valuable. However the most urgent need is for a treatment of the dynamics of the fluorescence in the diffusion regime which is better founded than our heuristic modification of the t matrix.

Note added in proof. Recent analysis shows that in a one-dimensional system with $X/W = \infty$ and $c_A \ll 1$, $f(t)$ decays asymptotically as $t^{1/2}\exp[-(c_A^2 t/t_0)^{1/3}]$ [B. Movaghar (private communication)].

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[5]The approached outlined in Ref. 4 involved a nonstandard form for $G_0(s)$. We have since repeated the calculation with the expression

$$G_0(s) = \int_0^\infty \exp\{-[s + X_{\mathrm{CPA}}(s)]t\} \times \exp[-(4/3)\pi^{3/2}2^{-1/2}n_D\beta^{1/2}t^{1/2}]\,dt \ ,$$

which is consistent with the definition used in this paper, and have obtained results for $X_{\mathrm{CPA}}(0)$ which differ by no more than about 10% from the values given previously. Similar results are obtained using the Green's function in the three-body approximation given by Gochanour *et al.* [C. R. Gochanour, H. C. Anderson, and M. D. Fayer, J. Chem. Phys. 70, 4254 (1979)].
[6]For mathematical details pertaining to the evaluation of $g_0(s)$ see K. K. Ghosh, thesis (University of Wisconsin, 1980) (unpublished).
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