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A Method of Calculation for an Infinite Continued Fraction —A particular reference to static limit of exciton migration—

Itsuko SATO*

The Doctoral Research Course in Human Culture, Ochanomizu University, Bunkyo-ku, Tokyo 112
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In a stochastic model of exciton transfer, a method of dynamical coherent potential approximation (DCPA) is useful. In this treatment, a propagator (Green's function) is expressed in terms of continued fraction.

In the present paper, a method of calculating the infinite continued fraction is given.

The method of DCPA gives very acculate results even in the static limit compared with the conventional static CPA. Thus DCPA can treat the static phenomena as well as the dynamical properties.

§ 1. Introduction

Sometimes we must treat problems of certain physical systems in a fluctuating environment. For instance, when an exciton propagates in a crystal, lattice viblations give rather profound effects destroying a phase coherence of the exciton[1]. In treating optical properties of excitons, a useful method of DCPA was employ by several authors[2),3)] and extended by us[4)].

Quite complicated interactions of excitons with the environment have been treated rather simply by introducing a stochastic model: The Hamiltonian is of the form

$$\mathscr{H}(t)=\mathscr{H}_0+\mathscr{H}_1(t) \quad , \tag{1.1}$$

$$\begin{aligned}\mathscr{H}_0 &= \sum_{\langle m,n\rangle} J_{mn}\, b_m^+\, b_n \\ &= \sum_k \omega_k\, a_k^+\, a_k \quad ,\end{aligned} \tag{1.2}$$

$$\mathscr{H}_1(t) = \sum_n \Delta_n(t)\ b_n^+\ b_n \tag{1.3}$$

*) Present address: Toshiba R&D Center, Toshiba Corporation, Komukai, Saiwai-ku, Kawasaki 210.

where $\mathscr{H}_0$ represents the coherent motion of excitons and J_{mn} is the transfer integral between site m and site n. We assume a nearest-neighbor interaction in (1.3), namely, J_{mn} takes the value J. We also put

$$\omega_k = \frac{J}{2} \sum_{\delta} e^{-i k \cdot \delta} \tag{1.4}$$

where δ represents a vector which connects a single site to its nearest neighbor site position. Moreover b_m^+, b_n are the exciton creation and anihilation operators in the site representation, and a_k^+, a_k are the corresponding Fourier transformed operators.

The coherent motion of excitons described by (1.2) are largely affected by the presence of $\mathscr{H}_1(t)$, where $\Delta_n(t)$ is a random function of time and their process are assumed to be the Gaussian Markoffian. According to the assumption, correlations of $\Delta_n(t)$'s are determined by

$$< \Delta_n(t) >_B = 0 \quad , \tag{1.5}$$

$$< \Delta_m(t) \, \Delta_n(t_1) >_B = \delta_{mn} \, \Delta_0^2 \, e^{-\gamma_0 |t - t_1|} \quad , \tag{1.6}$$

$$< \Delta_n(t) \, \Delta_n(t_1) \cdots \Delta_n(t_{j-1}) >_{B,C} = 0 \quad (\text{for } j \geqq 3) \, . \tag{1.7}$$

where the subscript "C" in (1.7) denotes the cumulant. The relation (1.6) represents the correlation of $\Delta_n(t)$'s; Δ_0 is a measure of strength of the fluctuation, γ_0 the decay rate of the correlation. When γ_0 tends to zero, the correlation time γ_0^{-1} becomes infinite, so that $\Delta_n(t)$ at a definite site n takes a certain constant value (static limit).

To discuss the optical absorption in the exciton problem, we need the propagator or Green's function. In the framework of these quantities are solved in the form of infinite continued fraction. In actual numerical calculations, it is necessary to truncate the continued fraction at an appropriate order. We present here a method to calculate these quantities and also discuss the behaviors in the static limit.

In the following we show the result of DCPA, and make clear the relation between the static limit of DCPA and the usual static CPA (Sec. 2).

Next we present a method to calculate the continued fraction, and discuss its convergent properties especially in the parameter region of the static limit (Sec. 3).

We give the conclusion in Section 4.

§ 2. The problem of exciton migration

2-1. The result of DCPA[2)–4)]

According to the DCPA method, the propagator (or Green's function) is given by

$$G(\omega) = \frac{1}{N} \sum_{k} G_k(\omega) \tag{2.1}$$

where

$$G_k(\omega) = \frac{1}{\omega - \omega_k - \Sigma_{01}(\omega)} \tag{2.2}$$

and

$$\Sigma_{0n}(\omega) = \frac{n\,\Delta_0^2}{F_0(\omega - \mathrm{i}\,n\,\gamma_0)^{-1} - \Sigma_{0n+1}(\omega)} \qquad (n \geqq 1) \quad , \tag{2.3}$$

$$F_0(\omega)^{-1} = G(\omega)^{-1} + \Sigma_{01}(\omega) \quad . \tag{2.4}$$

The self-energy $\Sigma_{01}(\omega)$ in (2.2) has been given by the recurrence relation (2.3), that is, we have the infinite continued fraction; the number n taking from 1 to infinity:

$$\Sigma_{01}(\omega) = \cfrac{\Delta_0^2}{F_0(\omega - \mathrm{i}\,\gamma_0)^{-1} - \cfrac{2\,\Delta_0^2}{F_0(\omega - 2\mathrm{i}\,\gamma_0)^{-1} - \cfrac{3\,\Delta_0^2}{F_0(\omega - 3\mathrm{i}\,\gamma_0)^{-1} - \cfrac{4\,\Delta_0^2}{\ddots}}}} \quad . \tag{2.5}$$

2-2. Continued fraction in the static problem and its integral representation.

In contrast to the result in Sec. 2-1., Sumi[5)] formulated a theory on the ground of the usual static CPA where the random potential is assumed to be the Gaussian distribution. In this case the $\Delta_n(t)$ in (1.3) is independent of t which corresponds to $\gamma_0 \equiv 0$. His result is

$$\Sigma_{01}(\omega)=\cfrac{\Delta_0^2}{F_0(\omega-\mathrm{i}\,\delta)^{-1}-\cfrac{2\,\Delta_0^2}{F_0(\omega-\mathrm{i}\,\delta)^{-1}-\cfrac{3\,\Delta_0^2}{F_0(\omega-\mathrm{i}\,\delta)^{-1}-\cfrac{4\,\Delta_0^2}{\ddots}}}}$$

$$(\omega;\ \text{real},\ \delta\to+0) \tag{2.6}$$

where δ is a convergent factor. The expression (2.5) coincides with (2.6) if we put $\gamma_0\equiv 0$. This fact has been confirmed by Sumi himself[2]. This is shown by a relation[6]

$$\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{e^{-t^2}}{z-t}\,\mathrm{d}t=\cfrac{1}{z-\cfrac{1/2}{z-\cfrac{2/2}{z-\cfrac{3/2}{\ddots}}}}\ . \tag{2.7}$$

If we use (2.7) then (2.1), (2.2) and (2.6) are written by

$$G(\omega)=\frac{1}{\sqrt{2\pi}\ \Delta_0}\int_{-\infty}^{\infty}\mathrm{d}\varsigma\ \frac{e^{-\varsigma^2/2\Delta_0^2}}{F_0(\omega)^{-1}-\varsigma}\ . \tag{2.8}$$

On the ground of the relation (2.4), (2.8) becomes

$$\frac{1}{\sqrt{2\pi}\ \Delta_0}\int_{-\infty}^{\infty}\mathrm{d}\varsigma\ e^{-\varsigma^2/2\Delta_0^2}\ T(\omega,\varsigma)=0\ , \tag{2.9}$$

$$T(\omega,\varsigma)=\frac{\varsigma-\Sigma_{01}(\omega)}{1-(\varsigma-\Sigma_{01}(\omega))\,G(\omega)} \tag{2.10}$$

and thus these equations satisfy the usual condition of the static CPA; "average of T-matrix is equal to zero". In (2.9)

$$\frac{1}{\sqrt{2\pi}\ \Delta_0}\ e^{-\varsigma^2/2\,\Delta_0^2}$$

represent a probability of the scattering by the potential.

§ 3. Numerical calculation

3-1. The method for calculating the infinite continued fraction

In 2-1. we derived $G(\omega)$ and $G_k(\omega)$ from with we have an absorption spectrum $I(\omega)$:

$$I(\omega) = \mathit{Im}\ G_{k=0}(\omega)\,. \tag{3.1}$$

The spectrum line shape is characterized by two parameters $\alpha_0 (\equiv \Delta_0/\gamma_0)$ and $\alpha' (\equiv \Delta_0/B)$ where $2B$ is the original band width of excitons.

When the modulation speed is fast ($\alpha_0 << 1$ and/or $\alpha' << 1$) the usual motional narrowing occurs. Convergence of the continued fraction is very fast; practically we have only to calculate only the lowest order term.

In an intermediate stage ($\alpha_0 \simeq 1$ and $\alpha' \simeq 1$), we must calculate the continued fraction up to $10 \sim 20$ orders.

While the modulation is slow (α_0, $\alpha' > 1$), we have to calculate up to very higher orders. This will be explained in more details in 3-2.

3-2. The results of numerical calculation and treatment of static limit

We give here several examples of calculations of the spectra in fig. 1 and fig. 2. In the static limit ($\gamma_0 \to 0$), we find the so-called Urbach tail:

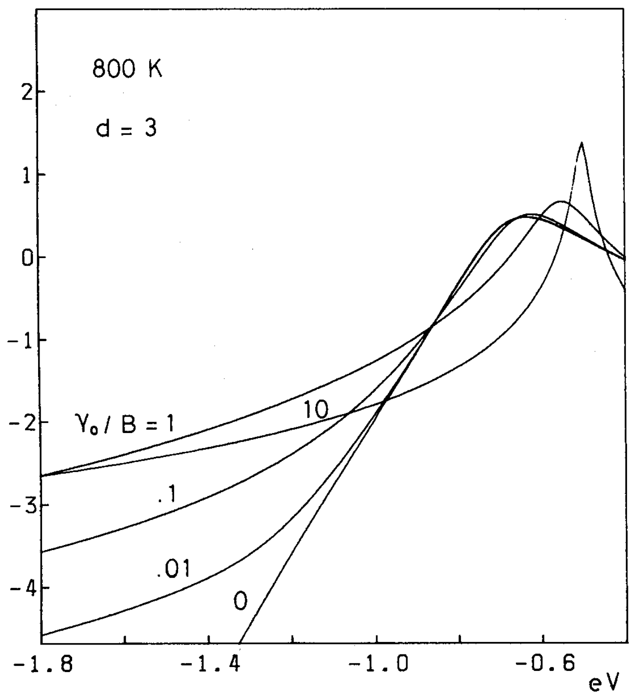

Fig. 1 Optical absorption spectra for the space dimension $d = 3$ at $T = 800\mathrm{K}$.

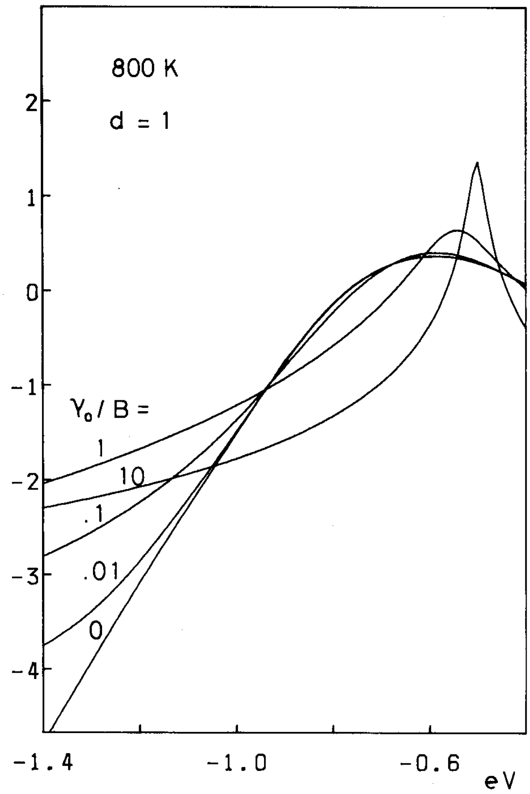

Fig. 2 Optical absorption spectra for the space dimension d = 1 at T = 800K.

$$e^{-A(\omega-\tilde{\omega})} \tag{3.2}$$

In the static limit, Schreiber and Toyozawa[7] made numerical simulations which were included together with our theoretical predictions in fig. 3 and fig. 4. The agreement of the theory with the simulation is almost perfect over the wide range of temperature and for any dimensions. In these figures, we used the relation

$$\Delta_0^2 = k_B T \tag{3.3}$$

following ref.7. We cannot obtain the Urbach tail in the usual perturbation theory. So it becomes clear that DCPA can be applied not only to the fast modulation case but also to the slow modulation case as is clear from fig.3 and fig.4.

In these calculations we solved (2.2), (2.5) making γ_0 tend to zero. On the other hand, in the Sumi's calculation based on the expression of the static CPA, we cannot fit the numerical result with the simulation data by Schreiber and Toyozawa[7] quantitatively. The fact is considered as follows. The numerical calculations done by Sumi[5] are based on an approximate

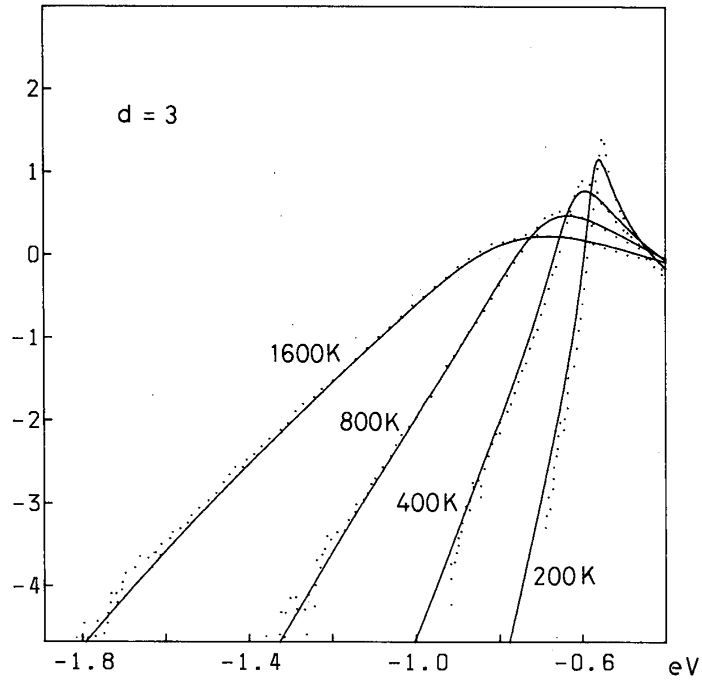

Fig. 3 Optical absorption spectra in the static limit for d = 3. The solid lines represent our theoretical results (based on the expression (2.5)) while the dots are taken from ref. 7.

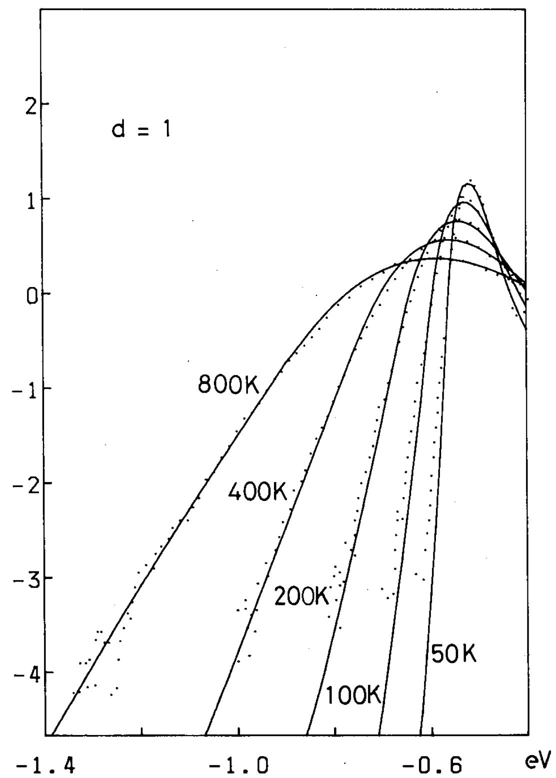

Fig. 4 Optical absorption spectra in the static limit for d = 1. The solid lines represent our theoretical results (based on the expression (2.5)) while the dots are taken from ref. 7.

expression [see (3.8) and figure caption of fig.4 in ref.5], or rather in his expression the equation satisfied by self-energy $\Sigma_0(\omega)$ or propagator $G(\omega)$ is written in a form of an integral equation and it may be a tedious task to obtain the sufficiently convergent value of $\Sigma_0(\omega)$ due to the difficulty inherent to the numerical calculations.

But if we use our representation (2.5), we have only to calculate the infinite continued fraction making γ_0 to vanish and truncating it at a certain sufficiently convergent order. For example, in fig.3 and fig.4 the convergent value of $\Sigma_0(\omega)$ is obtained at $\gamma_0/B = 10^{-3} \sim 10^{-4}$ with the order of 3000. We then identify these quantities as the optical absorption spectrum in the static limit.

Indeed Sumi[2] himself calculated the optical absorption spectrum with the use of (2.5), in which although the smallest value of γ_0/B is 10^{-2}, and he obtained the Urbach-like behavior. His result[2] is in fact more satisfactory than the previous ones[5]. However, the value of $\gamma_0/B(= 10^{-2})$ is still too large to regard the static limit behavior. In our calculation we make γ_0/B smaller ($10^{-3} \sim 10^{-4}$) and find the convergent values at a suitable order at which we truncate the infinite continued fraction. This is considered to be the static limit. As a result we could get an excellent agreement with the simulation data by Schreiber and Toyozawa[7].

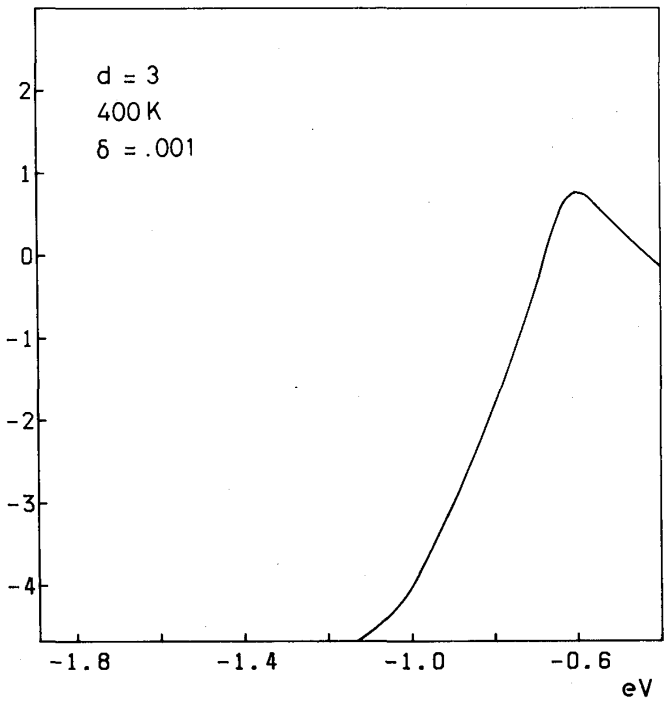

Fig. 5 Optical absorption spectrum in the static limit for $d = 3$ (based on the expression (2.6)).

On the other hand, we can also make a numerical calculation by means of the expression (2.6), but the convergent property of the infinite continued fraction is not satisfactory. For the comparison's sake, we give also a calculation using (2.6). For example, when T=400K, we have to consider the continued fraction up to 10^5 order to obtain the convergent value for $\delta=10^{-3}$. Moreover, even for $\delta=10^{-3}$ the spectrum is out of linear property (fig.5), so that the convergent property is quite bad. If we use (2.5), for $\gamma_0/B=10^{-4}$, the convergent order of continued fraction is 10^3, so that expression (2.5) is superior to (2.6).

§4. Conclusion

In this work we discussed a method of the numerical calculation of the infinite continued fraction.

First it is necessary to truncate it at a suitable order. This order is determined by the Kubo numbers α_0, α'. The standard truncating order ranges from 2 to 20. But in the slow modulation case, it becomes order of $10^2 \sim 10^3$ which is equal to $B/\gamma_0 (= \alpha_0/\alpha')$.

Moreover, we give a precise discussion about the numerical treatment in the slow modulation case together with the several analytic expressions in the static case. These are summarized as follows:
(i) Putting $\gamma_0 \equiv 0$ formally in (2.5), we obtain the result of static CPA by Sumi[5], (2.6).
(ii) On the other hand, the expression (2.5) with $\gamma_0 \to +0$ is considered to correspond to (2.6), but in the actual numerical treatments, (2.5) has a very rapid convergent property of the continued fraction than (2.6).
(iii) Consequently, it is concluded that the expression of our formulation of the DCPA is superior to that of the static CPA, not only in the treatment of the dynamical behavior of the absorption spectrum and density of state but also in the Urbach tail problem.

In this paper we discussed on the infinite continued fraction on the basis of the result of DCPA in the problem of exciton migration. But the method itself is not confined to the specific problem discussed here: In an extended theory of exciton migration, we obtained the more complicated infinite continued fraction[4]. Moreover in the theory of the low field resonance[8], we solved the problem exactly in certain cases using the similar treatment and obtained absorption spectrum in a form of the infinite continued fraction. In the numerical calculations of the infinite continued fraction we have some delicate problems. This will be discussed in the following paper.

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Appendix Programs list

We present here the programs list to calculate (2.5). In this program first we make an input of the parameters γ_0/B and Δ_0/B. Next we assign the energy region: an initial value and a final value of ω/B and division number are determined. Finally we give the truncating order and the number of iteration.

When (2.6) is calculated, we should put $K=1$ in 41 of the following list.

```
 H201

 1 C****GAUSSIAN(CPA METHOD)****DIAGONAL
 2 C                    M,N-TH APPROXIMATION
 3 C
 4 C****ABSORPTION AND DENSITY OF STATES****
 5 C
 6          COMPLEX CP1,CP,E1,Z
 7       *    ,DIM,DS
 8       *    ,CARD
 9 C
10          CABS(Z)=SQRT(REAL(Z)**2+AIMAG(Z)**2)
11 C
12          WRITE(2,100)
13 100      FORMAT(1H ,'R0,D0=')
14          READ(1,101) R0,D0
15 101      FORMAT(2F10.0)
16          WRITE(2,200)
17 200      FORMAT(1H ,'E0,EF,POINT=')
18          READ(1,201) E0,EF,IFF
19 201      FORMAT(2F10.0,I3)
20          WRITE(2,300)
21 300      FORMAT(1H ,'APPROXIMATION ORDER(M,N)=')
22          READ(1,301) MA,NA
23 301      FORMAT(2I10)
24 C
25          R1=0.
26          D1=0.
27          EH=(EF-E0)/(IFF-1)
28          M1=MA+1
29          N1=NA+1
```

```
C
      REWIND 5
      WRITE(5) R0,R1,D0,D1,IFF
      DO 40 J=1,IFF
      E=E0+EH*(J-1)
C
      CP1=CMPLX(0.,0.)
      DO 30 N=1,N1
      CP=CMPLX(0.,0.)
      DO 20 M=1,M1
      K=M1+1-M
      E1=CMPLX(E,-R0*K)
      Z=E1-CP1
      CP=D0*D0*K/(1./CARD(Z)+CP1-CP)
20    CONTINUE
      Z=CP-CP1
      IF(CABS(Z).LT.1.E-5) GO TO 1
      CP1=CP
30    CONTINUE
C
1     DIM=1./(E+1.-CP)
      GIM=AIMAG(DIM)
      Z=E-CP
      DS=CARD(Z)
      DD=AIMAG(DS)
C
      WRITE(2,400) E,GIM,DD,N
400   FORMAT(1H ,F6.3,2(2X,E17.9),2X,I3)
      WRITE(5) E,GIM,DD
40    CONTINUE
      ENDFILE 5
C
      STOP
      END
```

```
      COMPLEX FUNCTION CARD(Z)
      COMPLEX CARD,Z,YSQ
      YSQ=CSQRT(Z*Z-1.)
      YIM=AIMAG(YSQ)
      IF(YIM.GT.0.) YSQ=-YSQ
      CARD=(Z-YSQ)*2
      RETURN
      END
```

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