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Statistical model of relaxation of highly non-equilibrium persistent photoconductivity in inhomogeneous semiconductors

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Abstract. The theory of relaxation of highly non-equilibrium persistent photoconductivity in inhomogeneous semiconductor material has been developed. For a wide range of current carrier concentrations, the influence of the statistical properties of stochastic electrostatic potential reliefs on the kinetics of persistent conductivity has been investigated analytically and numerically. The predicted temporal dependence of the instantaneous relaxation time is consistent with the results of known experiments performed far from equilibrium and may be used for their interpretation.

At low temperature many semiconductor structures, especially compounds and semiconductor alloys, exhibit persistent conductivity after photo-excitation has ceased [1, 2]. This phenomenon is characterised by the extremely long lifetime of induced conductivity for rather high carrier concentrations. Persistent conductivity has been observed in experiments performed on $Al_xGa_{1-x}As$ [3], ZnSe [4, 5], GaAs [6], InP [7], Si [8] and others. To explain the effect, assuming homogeneity in the semiconductor several so-called 'centre models' were proposed, which attribute the prolonged kinetics to large lattice relaxation (DX centres) [9, 10] or to the excess carrier separation in $\boldsymbol{k}$-space [11, 12]. Another possible restriction on the recombination rate may be given by electrostatic 'collective barriers' produced by inhomogeneities in the material [2, 6, 13].

An abnormal relaxation of resistivity (ρ) was frequently registered experimentally after photo-excitation of the semiconductor, when the instantaneous relaxation time ($t_i = \rho/(\mathrm{d}\rho/\mathrm{d}t)$) grows linearly with time t

$$t_i = \gamma^{-1} t + \alpha \tag{1}$$

where γ and α are constants during the relaxation [2, 5, 12, 14, 15].

Such behaviour of t_i while ρ changes by two to three orders of magnitude and γ is essentially other than unity, can hardly be explained in terms of centre models.

Two approaches for interpreting the dependence (1) in the collective barriers model are known. In the first approach [14], the recombination (U_r) and drift (U_d) barriers are supposed to be constant and proportional to each other throughout the sample ($U_d < U_r$). In the second [15, 16], the much more realistic stochastic description of potential is considered, but only a small deviation of ρ from the equilibrium value is assumed.

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This paper develops the statistical model of relaxation of persistent photoconductivity, suitable for describing in a unified way both the highly non-equilibrium and near-equilibrium stages of relaxation in inhomogeneous semiconductors. The influence of statistical properties of the stochastic potential relief on the temporal kinetics of persistent conductivity is investigated analytically and numerically over a wide range of current carrier concentration.

Let us consider a semiconductor material with a spatially inhomogeneous stochastic electrostatic potential relief, generated by an inhomogeneous distribution of deep impurity centres, centre agglomerates or structure defects.

Here we will restrict ourselves to large-scale relief, when carrier tunnelling and a spatial dependence of their mobilities may be ignored. In this case the electrostatic fields cause only a spatial modulation of the bottom of the conduction band.

The electrons are assumed to be produced by photo-ionisation of deep impurity centres and to be thermalised in the conduction band. We shall consider the relaxation of the electron density after photo-excitation has ceased. It is because thermalised electrons accumulate in the potential relief minima, that defects are occupied by electrons here at the initial relaxation stages. Later, the main contribution in recombination rate will be given by the domains with the increasingly high electrostatic potential, because only there will the recharged defects remain. The recombination rate thus decelerates and will be determined by the properties of the 'tail' of the random potential relief distribution.

Denoting the concentration of defects able to capture electrons from the conduction band, in the forbidden gap at point $\boldsymbol{r}$ as $p(\boldsymbol{r}, t)$, and the conduction band electron concentration as $n(\boldsymbol{r}, t)$, we have

$$\mathrm{d}p(\boldsymbol{r}, t)/\mathrm{d}t = -Rn(\boldsymbol{r}, t)p(\boldsymbol{r}, t) \tag{2}$$

where R is the recombination coefficient.

For a non-degenerate electron gas

$$n(\boldsymbol{r}, t) = N(t)Z^{-1}(T)\exp[-\varepsilon(\boldsymbol{r})/kT] \tag{3}$$

where $N(t)$ is the average conduction band electron concentration, $\varepsilon(\boldsymbol{r})$ is the magnitude of the local random electrostatic potential, which defines the conduction band bottom at point $\boldsymbol{r}$, k is Boltzmann's constant, T is the temperature and $Z(T)$ is the partition function defined as

$$Z(T) = \langle\exp(-\varepsilon(\boldsymbol{r})/kT)\rangle \tag{4}$$

where $\langle \ldots \rangle$ indicates spatial averaging.

The electroneutrality condition connects the average concentration of free electrons to the average concentration of defects $P(t) = \langle p(\boldsymbol{r}, t)\rangle$:

$$N(t) = P(t) - P_0 \tag{5}$$

where P_0 is the equilibrium defect concentration.

Since, according to (2) and (3), the local recombination rate depends on the value of local electrostatic potential $\varepsilon(\boldsymbol{r})$, it is convenient to introduce the variable

$$p(\varepsilon, t) = \langle p(\boldsymbol{r}, t)\delta(\varepsilon - \varepsilon(\boldsymbol{r}))\rangle, \tag{6}$$

where $p(\varepsilon, t)$ is the average defect concentration in the domains where the potential belongs to the interval ε, $\varepsilon + \mathrm{d}\varepsilon$. It is obvious that

$$P(t) = \int p(\varepsilon, t)\,\mathrm{d}\varepsilon. \tag{7}$$

Multiplying equation (2) by $\delta(\varepsilon - \varepsilon(\boldsymbol{r}))$ and averaging both parts of the identity obtained over the $\boldsymbol{r}$ we have

$$\mathrm{d}p(\varepsilon, t)/\mathrm{d}t = -RN(t)Z^{-1}(T)\,\mathrm{e}^{-\varepsilon/kT}\,p(\varepsilon, t). \tag{8}$$

The formal solution of equation (8), expressed by the unknown function $N(t)$, takes the form

$$p(\varepsilon, t) = p(\varepsilon, 0)\exp\left(-[R\,\mathrm{e}^{-\varepsilon/kT}/Z(T)]\int_0^t N(t')\,\mathrm{d}t'\right). \tag{9}$$

In the case of uniformly spatially distributed defects, which generate electrons, the value of $p(\varepsilon, 0)$ is determined by the statistical distribution $F(\varepsilon)$ of the stochastic potential relief

$$p(\varepsilon, 0) = P(0)F(\varepsilon) \tag{10}$$

where

$$F(\varepsilon) = \langle\delta(\varepsilon - \varepsilon(\boldsymbol{r}))\rangle.$$

In this paper we consider the case where the photo-excited electron concentration may considerably exceed the equilibrium defect concentration, but is insufficient to change the electrostatic potential relief significantly. As the conductivity is determined by the electron concentration at the percolation level [17], in this case it is proportional to $N(t)$.

Substituting expression (5) into (7), taking into account (9) and (10), we get the self-consistent non-linear equation for the photo-excited electron average concentration $N(t)$

$$N(t) = \int \mathrm{d}\varepsilon\,(N(0) + P_0)F(\varepsilon)\exp\left(-RZ^{-1}(T)\,\mathrm{e}^{-\varepsilon/kT}\int_0^t N(t')\,\mathrm{d}t'\right) - P_0. \tag{11}$$

Introducing the dimensionless variables

$$I = RZ^{-1}(T)\int_0^t N(t')\,\mathrm{d}t' \tag{12}$$

$$Q = P_0[P_0 + N(0)]^{-1} \tag{13}$$

$$\tau = RZ^{-1}(T)(N(0) + P_0)t \tag{14}$$

we obtain from (11) the equation

$$\frac{\mathrm{d}I}{\mathrm{d}\tau} = \int \mathrm{d}\varepsilon\,F(\varepsilon)\exp[-I\exp(-\varepsilon/kT)] - Q \tag{15}$$

with the initial condition $I(0) = 0$.

It should be noted that an n-type semiconductor with equilibrium free electron concentration N_0 can be described by (15), provided that P_0 has been replaced by $-N_0$ in (5), (11), (13) and (14), and hence Q has become negative.

The value for γ in equation (1), being the subject of experimental investigation, can be directly expressed in terms of the variable I, introduced above, by

$$t_i = [N(t) - N_0]/|d(N(t) - N_0)/dt| = (N(t) - N_0)/dt|dN/dt|^{-1} \tag{16}$$

$$\gamma^{-1} = dt_i/dt = d[(\dot{I} - N_0)|\ddot{I}|^{-1}]/dt \tag{17}$$

where N_0 equals zero for i- and p-type material N_0 can be neglected for highly non-equilibrium relaxation stages. Naturally, the introduction of γ by means of (17) can be fruitful only if γ changes more slowly with time than does N. We show below that this condition is typical for the model under consideration.

We now show how equation (15) determines the recombination kinetics in the absence of potential relief fluctuations ($F(\varepsilon) = \delta(\varepsilon)$). Equation (15) is then reduced to

$$dI/d\tau = \exp(-I) - Q \tag{18}$$

with the solution

$$\tau = -(1/Q)\ln\{[1 - Q\exp(I)]/(1 - Q)\}. \tag{19}$$

The highly non-equilibrium initial condition $N(0) \gg P_0$ implies $Q \ll 1$. Hence at the initial relaxation stage, when $Q\exp(I) \ll 1$, equation (18) yields

$$I = \ln(1 + \tau) \tag{20}$$

$$N \sim \dot{I} = (1 + \tau)^{-1} \tag{21}$$

as it should be in the bimolecular recombination regime. If $\tau \to \infty$, then $I \to I_s = -\ln Q$ and

$$I - I_s = -(1 - Q)\exp(-Q\tau) \tag{22}$$

$$N \sim \dot{I} = (1 - Q)Q\exp(-Q\tau). \tag{23}$$

These relations correspond to the linear recombination law at the far relaxation stage when the free-electron concentration is essentially less than the defect concentration. The smaller Q is, the later the transition from the bimolecular to the linear regime occurs. The relaxation conductivity instantaneous time takes the form

$$\tau_i = \tau + \tau_0 \qquad \tau_0 = \text{constant} \tag{24}$$

for the bimolecular recombination regime; for the linear regime it is given by

$$\tau_i = R^{-1}P_0^{-1}. \tag{25}$$

Note that bimolecular recombination formally corresponds to equation (1) with $\gamma = 1$ and linear recombination to equation (1) with $\gamma \to \infty$.

The long-term relaxation stage is determined by the asymptotic behaviour of $F(\varepsilon)$ in the energy region $\varepsilon \gg \varepsilon_0$, where ε_0 is the characteristic amplitude of $\varepsilon(r)$ fluctuations. In most cases the asymptotics of $F(\varepsilon)$ is described by

$$F(\varepsilon) = \beta\exp[-(\varepsilon/\varepsilon_0)^\lambda]. \tag{26}$$

For a Gaussian random potential $\lambda = 2$, whilst for doped semiconductors $\lambda = \frac{3}{2}$ [16–18].

In reference [15], the relaxation for weak deviations from equilibrium and $\lambda = 1$ was considered. In the following we shall restrict ourselves to distribution functions of the type given by equation (26) with the normalising factor

$$\beta^{-1} = \int \exp[-(\varepsilon/\varepsilon_0)^\lambda]\,\mathrm{d}\varepsilon.$$

It follows from equation (15) that the I grows monotonically with τ. Further, we consider the most interesting case of the highly non-equilibrium relaxation stage, when the term Q in equation (15) may be omitted. The processes of near-equilibrium relaxation in inhomogeneous material are analysed in the Appendix.

Introducing the variable $\kappa = \varepsilon/kT$, taking into account equation (26) and considering the highly nonequilibrium relaxation stages, we obtain

$$\mathrm{d}I/\mathrm{d}\tau = \beta(\varepsilon_0, \lambda)kT\mathcal{I} \tag{27}$$

where

$$\mathcal{I} = \int_0^\infty \exp\{-[(\kappa/\kappa_0)^\lambda + I\,\mathrm{e}^{-\kappa}]\}\,\mathrm{d}\kappa$$

$$\kappa_0 = \varepsilon_0/kT.$$

For large I, the main contribution to the integral originates from the vicinity of the extremum point κ_m of the function

$$A(\kappa) = (\kappa/\kappa_0)\lambda + I\exp(-\kappa) \tag{28}$$

i.e. κ_m is the root of the equation

$$A'(\kappa) = \lambda\kappa_0^{-\lambda}\kappa^{\lambda-1} - I\exp(-\kappa) = 0. \tag{29}$$

For $I \to \infty$

$$\kappa_m \sim \ln I. \tag{30}$$

Taking into account for $\lambda \neq 1$ only the leading term of the expansion of $A(\kappa)$ in $(\kappa - \kappa_m)/\kappa_m$, we obtain

$$\mathcal{I} \sim \exp[-A(\kappa_m)] \int_{-\kappa_m}^\infty \exp[-\delta(\mathrm{e}^{-z} - 1 + z)]\,\mathrm{d}z \tag{31}$$

where $z = \kappa - \kappa_m$ and $\delta = \lambda\kappa_0^{-\lambda}\kappa_m^{\lambda-1}$.

The main contribution to the integral in equation (31) is made by values of z less than $\delta^{-1} = \lambda^{-1}\kappa_0^\lambda\kappa_m^{1-\lambda}$. Hence the $A(\kappa)$ expansion in z/κ_m is justified for $\kappa_m \gg 1$. The integral in (31) equals

$$(\mathrm{e}/\delta)^\delta\gamma(\delta\exp\kappa_m, \delta)$$

where $\gamma(x, y)$ is the incomplete gamma function. Hence, asymptotically

$$\mathcal{I} \sim (\mathrm{e}/\delta)^\delta\Gamma(\delta)\exp[-A(\kappa_m)] \tag{32}$$

where $\Gamma(x)$ is the gamma function. For sufficiently large I, equation (32) can be reduced to

$$\mathcal{I} \sim (2\pi\kappa_0^\lambda/\kappa_m^{\lambda-1}\lambda)\exp[-A(\kappa_m)] \qquad \lambda > 1 \tag{33}$$

$$\mathcal{I} \sim (\kappa_0^\lambda\kappa_m^{1-\lambda}/\lambda)\exp[-A(\kappa_m)] \qquad \lambda < 1 \tag{34}$$

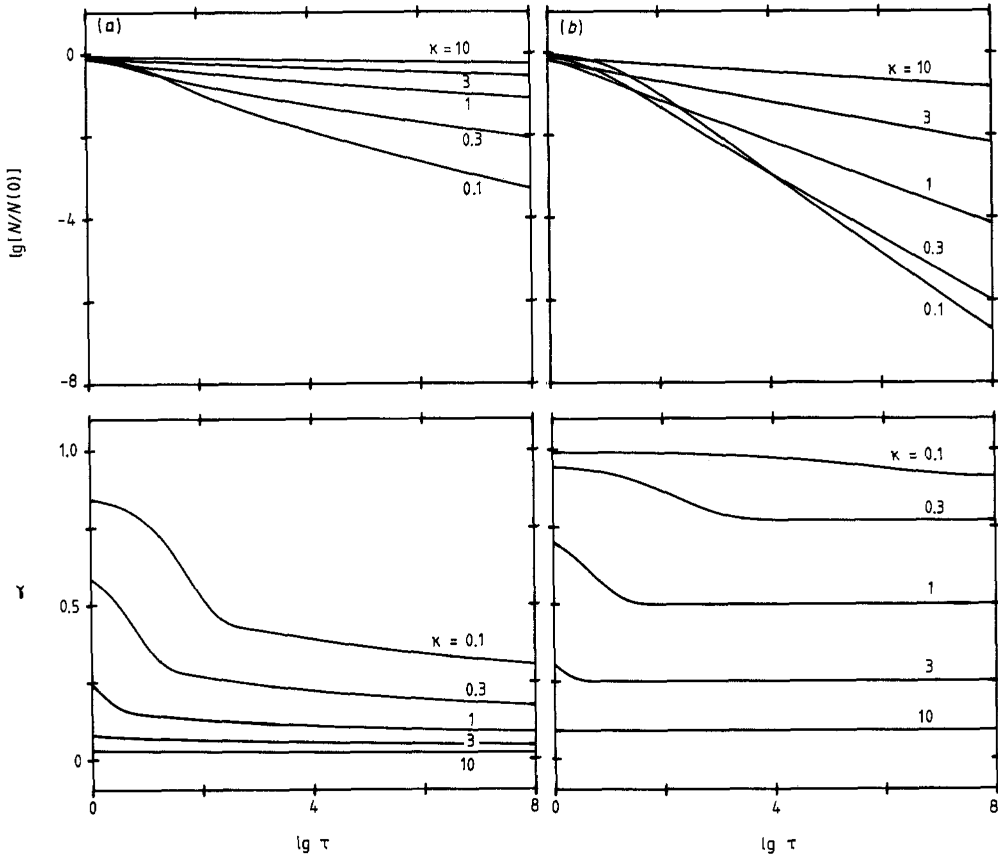

Figure 1. Temporal dependences of N and γ. (*a*) $\lambda = 0.5$, (*b*) $\lambda = 1$.

$$\mathcal{J} \sim (\mathrm{e}\kappa_0)^{1/\kappa_0}\Gamma(1/\kappa_0)I^{-1/\kappa_0} \qquad \lambda = 1. \tag{35}$$

In the last case

$$I \sim (\tau + \tau_0)^{\kappa_0/(1+\kappa_0)} \qquad \tau_0 = \text{constant} \tag{36}$$

$$N = \dot{I} \sim \kappa_0(1 + \kappa_0)^{-1}(\tau + \tau_0)^{-1/(1+\kappa_0)} \tag{37}$$

$$\gamma^{-1} = 1 + \kappa_0 = 1 + \varepsilon_0/kT \tag{38}$$

i.e. γ is a constant less than 1, the value of γ approaches unity with increasing temperature or with decreasing amplitude of the characteristic fluctuation of the potential relief.

The retention of only the leading term in (32) is sufficient for an asymptotic estimate of γ by means of equation (17)

$$\gamma^{-1} \sim 1 + \kappa_0^{\lambda}\lambda^{-1}(\ln I)^{1-\lambda}. \tag{39}$$

If $\lambda = 1$, then (39) implies $\gamma^{-1} = 1 + \kappa_0$ in agreement with (38).

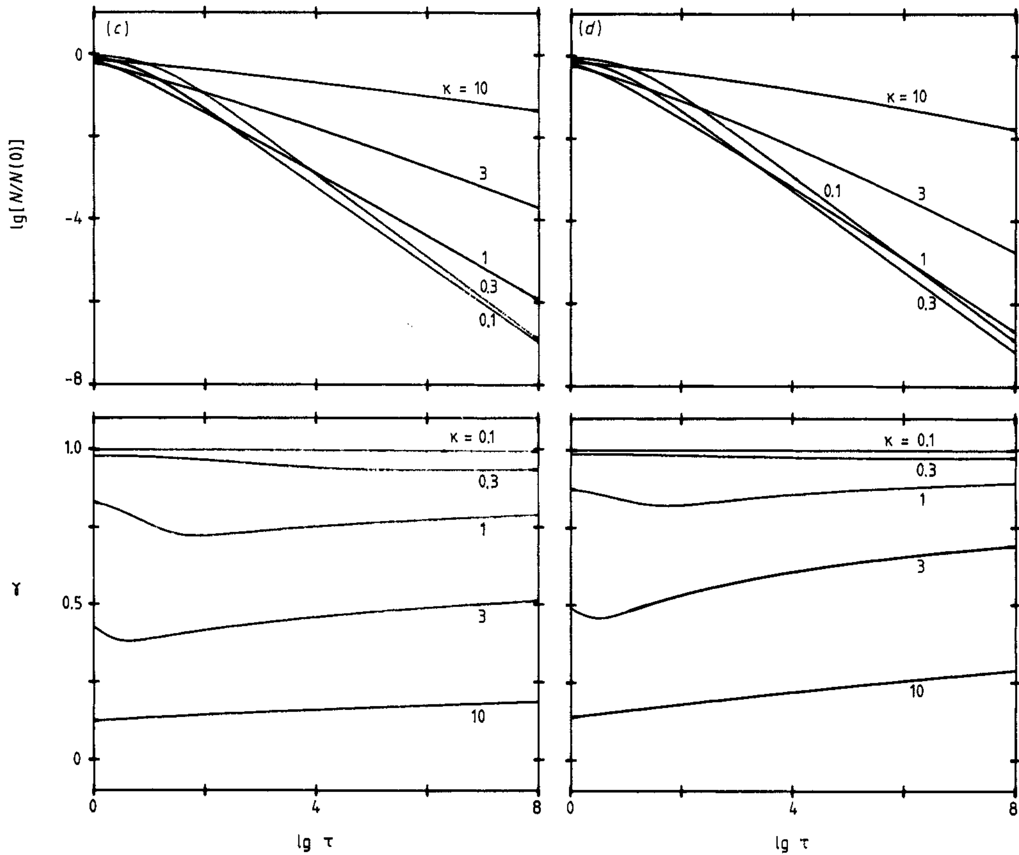

Figure 1 (continued). Temporal dependences of N and γ. (c) $\lambda = 1.5$, (d) $\lambda = 2$.

Equation (39) predicts qualitatively different asymptotic behaviour for γ as $t \to \infty$ for different values of λ:

$$\gamma \to 1 \qquad \lambda > 1$$

$$\gamma \to 0 \qquad \lambda < 1.$$

We have investigated in detail the relaxation behaviour of photo-excited current carriers for a wide range of electron concentrations by means of numerical computation of equation (27) for different parameters of the potential statistical distribution.

As shown in figures 1(a)–(d), extended temporal intervals always exist, during which the current carrier concentration changes by several orders of magnitude, but γ exhibits only weak variation, corresponding to the experimentally observed dependence (1). The long-term relaxation stage is well described by the analytical results obtained above.

Thus the model considered here provides a unified description of the temporal dynamics of the relaxation of conductivity in inhomogeneous semiconductor materials from the highly non-equilibrium stage to near-equilibrium. The solutions obtained are valid throughout a wide conductivity range and near the equilibrium are in agreement with the known results of references [15, 16].

The essential dependence of the instantaneous time t_1 of the conductivity relaxation on the statistical properties of the stochastic potential relief has been established. The discontinuous threshold character of limiting value of γ as a function of λ has been shown for the class of distribution function with asymptotes described by

$$F(\varepsilon) \sim \exp[-(\varepsilon/\varepsilon_0)^\lambda].$$

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Appendix

We have two cases near the equilibrium relaxation stage.

(i) $Q > 0$. Let us introduce the variable $x = I - I_s$ where I_s is the root of the right-hand side of equation (15). Now equation (15) may be rewritten as

$$\frac{dx}{d\tau} = \int d\varepsilon\, F(\varepsilon) \exp[-I_s \exp(-\varepsilon/kT)] \times \{\exp[-x \exp(-\varepsilon/kT)] - 1\}. \quad \text{(A1)}$$

If $x \ll 1$ then (A1) is reduced to

$$dx/d\tau = -\tau_{eq}^{-1} x \quad \text{(A2)}$$

with

$$\tau_{eq}^{-1} = \int \exp\{-[\varepsilon/kT + I_s \exp(-\varepsilon/kT)]\} F(\varepsilon)\, d\varepsilon.$$

Hence

$$x = A \exp(-\tau/\tau_{eq}) \qquad A = \text{constant}$$
$$\Delta N = N = dx/d\tau$$
$$\tau_i = \tau_{eq}$$
$$\gamma^{-1} = 0.$$

(ii) $Q < 0$ (n-type material). It is natural to introduce the variable $x = I - Q\tau$. It follows from (15) that

$$\frac{dx}{d\tau} \approx \int d\varepsilon\, F(\varepsilon) \exp[Q\tau \exp(-\varepsilon/kT)]. \quad \text{(A3)}$$

Using the above estimation we obtain

$$\Delta N = dx/d\tau \sim \beta kT(e/\delta)^\delta\, \Gamma(\delta) \exp[-A(\kappa_m)] \quad \text{(A4)}$$

where

$$A(\kappa_m) \sim \exp(-\kappa_m^{-\lambda} \ln^\lambda \tau).$$

Hence

$$\tau_i \sim \kappa_0^\lambda \tau/\lambda \ln^{\lambda-1} \tau \quad \text{(A5)}$$
$$\gamma^{-1} = \kappa_0/\lambda \ln^{\lambda-1} \tau. \quad \text{(A6)}$$

Equations (A5) and (A6) are in agreement with the results of references [15, 16].

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