

Theory of Kinetic Equations for Systems with Production and Absorption of Particles

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This paper is concerned with systems which are not in thermal equilibrium because of production and absorption of particles. On the basis of a new Hamiltonian describing such a nonequilibrium system, we develop a method for deriving kinetic equations for singlet density, time correlations of density, etc., including all the higher order interactions necessary to describe the production and absorption of particles. The foundations of the Boltzmann and Langevin equations for neutron distributions are studied. The time-correlation function is shown to obey a kinetic equation identical to that for the singlet density. It is also shown that the description of density fluctuations based on the Langevin equation is equivalent to the simplest decoupling of the rigorous hierarchy of equations for correlation functions.

KEY WORDS: Nonequilibrium systems; Boltzmann equation; Langevin equation; time correlations; systems with nuclear reactions.

1. INTRODUCTION

In recent years there has been a growing interest in systems which are not in thermal equilibrium even in stationary states because of the production and absorption of particles. Examples are systems with chemical reactions, partially ionized plasmas, various fusion devices, and nuclear fission reactors. A major problem in these nonequilibrium systems is to calculate phase-space

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distributions of the particles and space-time correlations of the particle density; they are usually discussed on the basis of phenomenological kinetic equations such as Boltzmann's transport equation and the Langevin equation.

The principal purpose of this paper is to develop a theory for the kinetic description of such nonequilibrium systems on the basis of quantum mechanical many-body theory. Although many first-principle approaches to various kinetic equations have been explored by many authors,⁽¹⁻⁴⁾ most of them are concerned with systems in which only scattering occurs, and which are, therefore, in thermal equilibrium in stationary states. Thus, very little is known about the foundations of the kinetic equations for the above-mentioned nonequilibrium systems.

Recent work of Girardeau⁽⁵⁾ is of interest in this respect. He developed a second-quantization representation for systems of atoms, ions, and electrons in such a way that composite particles (atoms and ions) are treated as elementary particles. He derived a Hamiltonian which takes explicit account of ionization and recombination processes. The Hamiltonian, therefore, seems to be very powerful for discussing, e.g., kinetic equations in partially ionized plasmas.

As a concrete example of the nonequilibrium systems, we consider in this paper a system of many neutrons, nuclei, and photons in nuclear reactors. Several authors have discussed the foundations and generalizations of the neutron transport equation, taking account of only scattering,^(6,7) or including the effects of nuclear reactions.⁽⁸⁾ In the latter formalism⁽⁸⁾ of Osborn *et al.*, however, the notion of the Hamiltonian is incomplete, and their deduction of various cross sections in kinetic equations is still phenomenological.

In previous papers^(9,10) we applied the Girardeau formalism and found the Hamiltonian to be given by Eq. (3) below, which describes nonequilibrium systems of neutrons, nuclei, and photons. On the basis of this Hamiltonian we shall study in this paper the foundations, validity, and generalizations of the usual kinetic equations.

We shall derive kinetic equations for neutron singlet density $F(x, t)$ and for the time-correlation function $F(xt; x't')$ of the neutron density. The resulting kinetic equation for the singlet density reduces to the usual one when quantum effects are neglected. It is found that the kinetic equation for the time-correlation function is identical to that for the singlet density, in the approximation of the simplest decoupling of a rigorous hierarchy of equations for correlation functions. This simplest decoupling is shown to be equivalent to the approximation underlying the Langevin equation.

In Section 2 we shall derive a rigorous hierarchy of equations for time-correlation functions. In Section 3 the simplest decoupling of the hierarchy is carried out to get the kinetic equation for $F(xt; x't')$. Section 4 is concerned

with the singlet density, and the final section is devoted to a summary and concluding remarks.

2. HIERARCHY OF EQUATIONS FOR TIME-CORRELATION FUNCTIONS

Let us consider many neutrons in a relatively dense medium consisting of nuclei U . The following reactions are assumed to occur in the system :

$$n + U \begin{cases} \rightarrow n + U & \text{(direct scattering)} \\ \rightarrow C \begin{cases} \rightarrow n + U & \text{(resonance scattering)} \\ \rightarrow \gamma + C & \text{(radiative capture)} \\ \rightarrow A + B \rightarrow n + a + B & \text{(fission)} \end{cases} \end{cases} \quad (1)$$

where n , C , and γ denote a neutron, a compound nucleus, and a photon, respectively. For simplicity we have assumed the simplest process for fission; the compound nucleus splits into two primary fragments A and B , one of which then emits only one prompt neutron. The generalization is given in the appendix.

By virtue of the ideal space introduced in Ref. 9 (hereafter Refs. 4, 7, 9, and 10 will be referred to as I, II, III, and IV, respectively), all the particles (neutron, nuclei, and photon) can be treated as elementary particles. A state $|U\rangle$ of nucleus U , for example, corresponds in the ideal space to the ideal state $|U\rangle$ which is created by the ideal operator U^\dagger . The creation operator of a neutron with wavenumber vector \mathbf{k} is denoted by $\eta_{\mathbf{k}}^\dagger$ and that of a photon with wavenumber vector \mathbf{q} and polarization λ is $b_{\mathbf{q}\lambda}^\dagger$. These ideal operators are defined by the simple commutation rules (III 2.11) for elementary bosons and fermions. The nuclear reactions in (1) are described² by a combination of the ideal Hamiltonians (IV 2.7) and (III 4.11). To discuss the transport of neutrons, however, we must specify the position and momentum of a neutron simultaneously, and we use here Ono's method of cell function.^(2,4,8) The configuration space of the neutrons is divided into cubic cells with edge length L , and states of a free neutron are specified by the phase points $x = (\mathbf{X}, \mathbf{P})$ instead of the wavenumber vectors. The edge length L is chosen to be of the order of magnitude of the spatial resolution with which we wish to describe the neutron distributions. The streaming of the neutrons is described by ^(4,8)

$$T = \sum_{\mathbf{x}, \mathbf{p}} \eta^\dagger(\mathbf{X}\mathbf{P}) \left(-\frac{i\hbar}{m} \mathbf{P} \cdot \nabla_{\mathbf{x}} \right) \eta(\mathbf{X}\mathbf{P}) \quad (2)$$

² To describe the last stage $A + B \rightarrow n + a + B$ in fission, we incorporate the open channels involving three particles n , a , and B into the argument of §2 in IV. We then find that the interaction potential $W(A; na)$ and the free Hamiltonian H_{0a} are added to (IV 2.7).

where m is the neutron mass and $\nabla_{\mathbf{x}}$ denotes a finite difference. Our system is thus characterized by the following Hamiltonian:

$$H = H_0 + T + H' \quad (3)$$

where H_0 is the free Hamiltonian

$$\begin{aligned} H_0 &= H_{0n} + H_{0U} + H_{0C} + H_{0A} + H_{0B} + H_{0a} + H_{0\gamma}; \\ H_{0n} &= \sum_{\mathbf{x}} E(\mathbf{P})\eta^\dagger(\mathbf{x})\eta(\mathbf{x}) \\ H_{0U} &= \sum_{\mathbf{U}} E(\mathbf{U})U^\dagger U, \quad \text{etc.}, \\ H_{0\gamma} &= \sum_{\mathbf{q}, \lambda} \hbar c q (b_{\mathbf{q}\lambda}^\dagger b_{\mathbf{q}\lambda} + \frac{1}{2}) \end{aligned} \quad (4)$$

and H' is the interaction potential

$$\begin{aligned} H' &= V + W, \quad W = W_1 + W_2, \\ W_1 &= W(C; nU) + W(C; AB) + W(C; \gamma C) \end{aligned} \quad (5)$$

$$V = \sum_{\mathbf{x}, \mathbf{U}} \sum_{\mathbf{x}', \mathbf{U}'} \eta^\dagger(\mathbf{x}) U^\dagger \langle \mathbf{x} U | v | \mathbf{x}' U' \rangle U' \eta(\mathbf{x}') \quad (6)$$

$$W(C; nU) = \sum_C \sum_{\mathbf{x}, \mathbf{U}} C^\dagger \langle C | H | \mathbf{x} U \rangle U \eta(\mathbf{x}) + \text{H.c.} \quad (7)$$

$$W(C; AB) = \sum_C \sum_{A, B} C^\dagger \langle C | H | AB \rangle BA + \text{H.c.} \quad (8)$$

$$W(C; \gamma C) = \sum_C \sum_{C', \mathbf{q}, \lambda} g(C, C', \mathbf{q}\lambda) C^\dagger C' (b_{\mathbf{q}\lambda} + b_{-\mathbf{q}\lambda}^\dagger) \quad (9)$$

$$W_2 = W(A; na) = \sum_{\mathbf{x}, a} \sum_A \eta^\dagger(\mathbf{x}) a^\dagger \langle \mathbf{x} a | H | A \rangle A + \text{H.c.} \quad (10)$$

where H.c. stands for the Hermitian conjugate. The interaction potential V describes the direct scattering; v in the matrix element is the hard-core potential for n - U interaction. The potential $W(C; nU)$ represents the resonance scattering and the first stage of the fission, $W(C; AB)$ represents the second stage, and $W(A; na)$ represents the last stage. The radiative capture is described by $W(C; \gamma C)$. This Hamiltonian is valid for neutrons in a dense medium, where n - U interaction is predominant. In general, we must take account of n - n and n - a interactions, etc., which will be ignored in this paper.

Since we are interested in the neutron density, we start with the Heisenberg equation of motion

$$i\hbar \frac{\partial}{\partial t} N_{\mathbf{H}}(x, t) = -\frac{i\hbar}{m} \mathbf{P} \cdot \nabla_{\mathbf{x}} N_{\mathbf{H}}(x, t) + H'_{x\mathbf{H}}(t) - H_{x\mathbf{H}}^\dagger(t) \quad (11)$$

for the number operator $N_{\mathbf{H}}(x, t) = \exp(iHt/\hbar) \eta^\dagger(x)\eta(x) \exp(-iHt/\hbar)$. In

(11), $H'_{xH}(t)$ is the Heisenberg operator corresponding to

$$\begin{aligned} H'_{x'} &= \eta^\dagger(x)[\eta(x), H'] \\ &= \sum_{U'} \sum_{v'} \eta^\dagger(x) U^\dagger \langle xU|v|x'U'\rangle U' \eta(x') \\ &\quad + \sum_{\bar{U}} \sum_{\bar{C}} \eta^\dagger(x) U^\dagger \langle xU|H|C\rangle C + \sum_{\bar{a}} \sum_{\bar{A}} \eta^\dagger(x) a^\dagger \langle xa|H|A\rangle A \end{aligned} \quad (12)$$

This expression is valid irrespective of whether the nuclei are fermions or bosons. We note here that the total number operator of the neutrons does not commute with the Hamiltonian; this is due to the interaction potential W and indicates that our system is in nonequilibrium.

The equation of motion (11) yields an equation for the time-correlation function $F(xt; x't')$ of the neutron density in the following way: Let the state of the system be denoted by $|\Psi_H\rangle$ in the Heisenberg picture. It corresponds in the ideal space to an ideal state $|\Psi_H\rangle$, as seen from III. Then the time-correlation function is expressed by

$$\begin{aligned} F(xt; x't') &= (1/L^6) (\Psi_H | N_H(x, t) N_H(x', t') | \Psi_H) \\ &= (1/L^6) \text{Tr} \{ \rho_H N_H(x, t) N_H(x', t') \} \end{aligned} \quad (13)$$

where $\rho_H = |\Psi_H\rangle\langle\Psi_H|$ is the density operator. Hence we have

$$(\partial/\partial t)F(xt; x't') = -(\mathbf{P}/m) \cdot \nabla_{\mathbf{x}} F(xt; x't') + C(xt; x't') \quad (14)$$

$$C(xt; x't') = (1/i\hbar L^6) \text{Tr} [\rho_H \{ H'_{xH}(t) - H'_{xH}(t) \} N_H(x', t')] \quad (15)$$

The function $C(xt; x't')$ contains the higher order correlation functions, and (14) is the first equation in a hierarchy of equations for correlation functions.

3. KINETIC EQUATION FOR THE TIME-CORRELATION FUNCTION

In this section we shall decouple the hierarchy of equations to get the kinetic equation (40) for the time-correlation function.

3.1. Decoupling of the Hierarchy

As a simple, but still useful approximation to $C(xt; x't')$, we express it in terms of the lowest order correlation function. Namely, we insert in (15) the identity operator $N_H^{-1}(x_0, t_0) N_H(x_0, t_0)$, and approximate the average of the product by the following product of averages:

$$C(xt; x't') \simeq \Gamma(xt; x_0 t_0) F(x_0 t_0; x't') \quad (16)$$

$$\Gamma(xt; x_0 t_0) = (1/i\hbar) \text{Tr} [\rho_H \{ H'_{xH}(t) - H'_{xH}(t) \} N_H^{-1}(x_0, t_0)] \quad (17)$$

Here x_0 and t_0 are to be determined later³; t_0 will be chosen to be the initial time of the transition described by $H'_{xH}(t)$ in (17). The insertion of the identity operator is only a mathematical device to simplify the discussion. Actually, the factor $N_H(x_0, t_0)$ arises from $H'_{xH}(t) - H'_{xH}(t_0)$, and hence the insertion of the identity operator is spurious. An approximation like (16), i.e., factorization of a higher order function into lower order ones, is often made in many-body theory, but its validity is not so clear and is subject to an examination of the final result. It will be seen later that (16) is a useful approximation if $t > t'$, and hence we shall assume $t > t'$ for a while.

Equation (14) is, however, still coupled because the function $\Gamma(xt; x_0t_0)$ in (16) is a higher order correlation function. We now express $\Gamma(xt; x_0t_0)$ in terms of the neutron singlet density by means of the adiabatic switching-off of interaction presented in I. This method is quite suitable for the present purpose, because it involves all the higher order interactions necessary to describe nuclear resonance reactions. The method is based on the view that the particles must be far apart before interaction, and that, therefore, the higher order densities can be expressed at the initial time of the interaction by products of singlet densities. It is by this procedure that we go over from the rigorous microscopic description to the irreversible kinetic description.

Let us consider that our system is now approaching a stationary state and make the following assumptions:

(a) The range of the neutron–nuclear interaction is so short that the neutrons are found in one of the free states, and the scattering and nuclear reactions occur instantaneously.

(b) The effects of the perturbations T , V , and W to this free state are separated.

(c) The transition probability for a time interval τ is proportional to τ .

The assumptions (a) and (b) are implications of the Boltzmann equation. The assumption (c) is used in Kirkwood's theory⁽¹⁾ and makes the rigorous equation irreversible⁽⁴⁾; $\tau \sim 10^{-14}$ sec is the time taken for a nuclear reaction to be completed, and the limit $\tau \rightarrow 0$ is taken at the end of the calculation in accordance with assumption (a).

According to assumption (a) the system is in a free state at an initial time t of a transition. Let this free state be denoted by $|I\rangle$, which is an eigenstate of the free Hamiltonian H_0 . Let us consider the time interval $t \sim t + \tau$ and employ the prescription of I for the time derivative. Then the rigorous

³ If the system is nonlinear, i.e., if n - n interaction is taken into account, the right-hand side of (16) should consist of two terms corresponding to two possibilities of determining x_0 .

equation (14) becomes, in the approximation (16),

$$\begin{aligned}
 & (\partial/\partial t)F(xt; x't') \\
 &= -(\mathbf{P}/m) \cdot \nabla_{\mathbf{x}} F(xt; x't') \\
 & \quad + \lim_{\tau \rightarrow 0} (1/\tau) \int_t^{t+\tau} \Gamma(x\bar{t}; x_0t) d\bar{t} F(x_0t; x't') \quad \text{for } t > t' \quad (18)
 \end{aligned}$$

where we have set $t_0 = t$. The coefficient of $F(x_0t; x't')$ in the second term on the right-hand side of (18) is identical in form with the collision term (I 3.18), except for the presence of $N_{\bar{H}}^{-1}(x_0, t)$; this extra factor, however, becomes a c -number when we rewrite $\Gamma(x\bar{t}; x_0t)$ in the interaction picture, as in (I 3.20). Hence, the calculation of $\Gamma(x\bar{t}; x_0t)$ is quite similar to that from (I 3.20) to (I 3.25). Namely, introducing the interaction picture, and switching off the interaction at the initial time, we obtain

$$\Gamma(x\bar{t}; x_0t) = \{C_1(x, t) + C_2(x, t)\}/F(x_0, t) \quad (19)$$

$$\begin{aligned}
 C_1(x, t) &= (1/i\hbar L^3) \text{Tr}\{\rho(t)T_x(0)\} + \text{c.c.} \\
 &= (1/i\hbar L^3) \langle I|T_x(0)|I \rangle + \text{c.c.} \quad (20)
 \end{aligned}$$

$$C_2(x, t) = (1/\hbar^2 L^3) \int_{-\infty}^0 e^{\epsilon t'/\hbar} \langle I|T^+(t')T_x(0)|I \rangle dt' + \text{c.c.} \quad (21)$$

Here, c.c. stands for the complex conjugate, and the time dependence of the operators is due to the interaction picture. In (19), $F(x_0; t) = \text{Tr}\{\rho(t)N(x_0, t)\}/L^3$ is the singlet density of the neutrons. The transition operators T and T_x are defined by

$$T = H'U(0, -\infty) \quad \text{and} \quad T_x = H'_x U(0, -\infty) \quad (22)$$

where $U(0, -\infty) = 1 + G(E_i)H' + \dots$ and $G(E_i) = (E_i - H_0 + i\epsilon)^{-1}$, ϵ being an infinitesimal positive number. They describe transitions of the many-particle state with the initial energy E_i ; the operator T_x represents all the transitions in which a neutron ends up in the state $|x\rangle$. It should be noted that the function $\Gamma(x\bar{t}; x_0t)$ has become independent of \bar{t} by virtue of the adiabatic-switching procedure. Hence the integration of Γ in (18) is proportional to τ [assumption (c)], and the limit $\tau \rightarrow 0$ in (18) has no effect; the second term on the right-hand side of (18) is thus actually the time average of the change of $F(xt; x't')$ caused by scattering and nuclear reactions (Kirkwood's time-averaging procedure⁽¹⁾). Since $|I\rangle$ is the free state, both $C_1(x, t)$ and $C_2(x, t)$ are given in terms of a product of the singlet densities, and thus (14) has become a closed equation.

3.2. Binary-Interaction Approximation

The transition operator T has nonvanishing matrix elements between states of more than two particles. Such matrix elements arise from higher

order interactions in which the colliding pair is different from the pair of the first collision. Since the transport equation implies that a neutron interacts with individual nuclei independently, we discard in this section such complicated interactions and assume that all the higher order interactions take place between the initially colliding pair. This approximation is expressed by the following diagrams:

$$T^{(V)} = V + VGV + VGVGV + \dots$$

$$\approx \dots \quad (23)$$

$$T^{(W_1)} = W_1 + W_1GW_1 + W_1GW_1GW_1 + \dots$$

$$\approx \dots \quad (24)$$

where $T = T^{(V)} + T^{(W)}$ according to assumption (b), and

$$T^{(W)} = T^{(W_1)} + T^{(W_2)} + T^{(W_2)}GT^{(W_1)} + \dots$$

Each internal line in the diagrams indicates the pairing of a creation operator and an annihilation operator in two successive interactions, which means that a particle destroyed in an interaction is the same as that created in the preceding interaction. In the case of the scattering, only the binary collision contributes in this approximation, and the effect of multiple scattering represented by

$$\dots \quad (25)$$

is ignored. This effect was examined in detail in II and will be considered in the present formalism in the appendix.

In the above-mentioned approximation we can write as follows:

$$T_x \approx \sum_U \sum_{x', U'} \eta^\dagger(x) U^\dagger(xU | T | x'U') U' \eta(x')$$

$$+ \sum_{a, B} \sum_{x', U'} \eta^\dagger(x) a^\dagger B^\dagger(xaB | T | x'U') U' \eta(x') \quad (26)$$

3.3. A Kinetic Equation for the Time-Correlation Function

Let us return to (19) and consider first the term $C_1(x, t)$. With the aid of the approximation (26), it readily follows that

$$(I | T_x(0) | I) = \sum_U (xU | T | xU) n(x, t) n(U, t) \quad (27)$$

where $n(x, t)$ and $n(U, t)$ are the occupation numbers in the state $|I\rangle = |\Psi(t)\rangle$. Substitution of (27) into (20) gives

$$\begin{aligned} C_1(x, t) &= \sum_{\bar{U}} \frac{2}{\hbar} \text{Im}(xU|T|xU) \frac{1}{L^3} n(x, t)n(U, t) \\ &= \sum_{\bar{U}} \left(-\frac{P}{m} \right) \sigma_{\text{tot}}(x, U) \frac{n(U, t)}{L^3} \frac{n(x, t)}{L^3} \end{aligned} \quad (28)$$

where use has been made of the optical theorem (III 5.26); $\sigma_{\text{tot}}(x, U)$ is the total cross section of nucleus U in the state $|U\rangle$. The sum of $\sigma_{\text{tot}}(x, U)n(U, t)$ with respect to U is the cross section of all U -type nuclei for a neutron in the \mathbf{X} cell; this cross section divided by the volume L^3 is the so-called macroscopic cross section $\Sigma_{\text{tot}}(x)$; here we have neglected the time dependence of Σ_{tot} because the total number of U -type nuclei is much greater than that of the neutrons. On the other hand, the factor $n(x, t)/L^3 = (I|N(x)|I)/L^3$ in (28) is the singlet density $F(x, t)$. Hence,

$$C_1(x, t) = -(P/m) \Sigma_{\text{tot}}(x)F(x, t) \quad (29)$$

which represents the number of neutrons with momentum \mathbf{P} "lost" in the \mathbf{X} cell per unit time and per unit volume.

Let us next consider the "gain" term $C_2(x, t)$, (21). By making use of the approximation (26), we readily obtain

$$\begin{aligned} &(I|T^+(t')T_x(0)|I) \\ &= \sum_{\bar{U}} \sum_{\mathbf{P}', \bar{U}'} \exp[-i(E(\mathbf{P}, U) - E(\mathbf{P}', U'))t'/\hbar] \\ &\quad \times |(\mathbf{XPU}|T|\mathbf{XP}'U')|^2 [1 \pm n(U, t)] \\ &\quad \times [1 - n(\mathbf{XP}, t)]n(U', t)n(\mathbf{XP}', t) \\ &+ \sum_{a, B} \sum_{\mathbf{P}', \bar{U}'} \exp[-i(E(\mathbf{P}, a, B) - E(\mathbf{P}', U'))t'/\hbar] \\ &\quad \times |(\mathbf{XP}aB|T|\mathbf{XP}'U')|^2 [1 \pm n(B, t)] \\ &\quad \times [1 \pm n(a, t)][1 - n(\mathbf{XP}, t)]n(U', t)n(\mathbf{XP}', t) \end{aligned} \quad (30)$$

Here, $E(\mathbf{P}, U) = E(\mathbf{P}) + E(U)$, etc.; the sign in front of the occupation numbers is determined by whether the nuclei are bosons or fermions; and use has been made of the fact that the transitions take place within a cell because of the short-range interaction. Inserting (30) into (21), and introducing the macroscopic differential cross sections by

$$\Sigma_s(\mathbf{XP}' \rightarrow \mathbf{XP}) = \sum_{\bar{U}, \bar{U}'} \frac{1}{L^3} \sigma_s(\mathbf{XP}'U' \rightarrow \mathbf{XP}U) [1 \pm n(U, t)]n(U', t) \quad (31)$$

$$\begin{aligned} \Sigma_f(\mathbf{XP}' \rightarrow \mathbf{XP}) &= \sum_{a, B, \bar{U}'} \frac{1}{L^3} \sigma_f(\mathbf{XP}'U' \rightarrow \mathbf{XP}aB) \\ &\quad \times [1 \pm n(B, t)][1 \pm n(a, t)]n(U', t) \end{aligned} \quad (32)$$

where, as was shown in III and IV,

$$\sigma_s(\mathbf{X}\mathbf{P}'U' \rightarrow \mathbf{X}\mathbf{P}U) = \frac{mL^3}{P'} \frac{2\pi}{\hbar} \delta(E(\mathbf{P}, U) - E(\mathbf{P}', U')) |(\mathbf{X}\mathbf{P}U|T|\mathbf{X}\mathbf{P}'U')|^2 \quad (33)$$

is the differential cross section for the direct and resonant scattering, and where

$$\sigma_f(\mathbf{X}\mathbf{P}'U' \rightarrow \mathbf{X}\mathbf{P}aB) = \frac{mL^3}{P'} \frac{2\pi}{\hbar} \delta(E(\mathbf{P}, a, B) - E(\mathbf{P}', U')) |(\mathbf{X}\mathbf{P}aB|T|\mathbf{X}\mathbf{P}'U')|^2. \quad (34)$$

is the fission cross section, we finally obtain

$$C_2(\mathbf{X}\mathbf{P}, t) = \sum_{\mathbf{P}''} \frac{P''}{m} \Sigma_s(\mathbf{X}\mathbf{P}'' \rightarrow \mathbf{X}\mathbf{P}) [1 - L^3 F(\mathbf{X}\mathbf{P}, t)] F(\mathbf{X}\mathbf{P}'', t) \\ + \sum_{\mathbf{P}''} \frac{P''}{m} \Sigma_f(\mathbf{X}\mathbf{P}'' \rightarrow \mathbf{X}\mathbf{P}) [1 - L^3 F(\mathbf{X}\mathbf{P}, t)] F(\mathbf{X}\mathbf{P}'', t) \quad (35)$$

which is the number of neutrons with momentum \mathbf{P} produced in the \mathbf{X} cell per unit time and per unit volume due to scattering and fission.

We are now in a position to combine (19), (29), and (35) with (18). Substitution of (19) into (18) gives the combination $\{F(x_0, t)\}^{-1} F(x_0 t; x' t')$, in which we can put x_0 arbitrarily. On putting $x_0 = x$ in the C_1 term in (19) and $\mathbf{X}_0 = \mathbf{X}$ and $\mathbf{P}_0 = \mathbf{P}''$ in the C_2 term, we arrive at the following kinetic equation:

$$\frac{\partial}{\partial t} F(xt; x' t') = -B(x)F(xt; x' t') \quad \text{for } t > t' \quad (36)$$

$$B(x)F(xt; x' t') = \frac{\mathbf{P}}{m} \cdot \nabla_{\mathbf{x}} F(xt; x' t') + \frac{P}{m} \Sigma_{\text{tot}}(x)F(xt; x' t') \\ - \sum_{\mathbf{P}''} \frac{P''}{m} \{\Sigma_s(\mathbf{X}\mathbf{P}'' \rightarrow \mathbf{X}\mathbf{P}) + \Sigma_f(\mathbf{X}\mathbf{P}'' \rightarrow \mathbf{X}\mathbf{P})\} \\ \times [1 - L^3 F(\mathbf{X}\mathbf{P}, t)] F(\mathbf{X}\mathbf{P}'' t; x' t') \quad (37)$$

Usually, the density correlation is discussed on the basis of the Langevin equation

$$(\partial/\partial t)\tilde{F}(x, t) = -B(x)\tilde{F}(x, t) + \tilde{R}(x, t) \quad (38)$$

for the stochastic distribution function $\tilde{F}(x, t)$, with the assumption that the power spectrum of the random force $\tilde{R}(x, t)$ is white, i.e.,

$$\langle \tilde{R}(x, t)\tilde{R}(x', t') \rangle \propto \delta(t - t')$$

If $t > t'$, we have the relation $\langle \tilde{R}(x, t)\tilde{F}(x', t') \rangle = 0$, and we easily find the kinetic equation (36) from the phenomenological equation (38). Hence we see that the rather drastic approximation (16) is useful only for $t > t'$. If

$t < t'$, on the other hand, we consider the derivative with respect to t' , and in a way quite similar to that in (36), we get

$$(\partial/\partial t')F(xt; x't') = -B(x')F(xt; x't') \quad \text{for } t < t' \quad (39)$$

The derivation of the kinetic equations (36) and (39) shows that the description of the fluctuations based on the Langevin equation (38) is interpreted as the approximation of the simplest decoupling of the rigorous equation (14), i.e., as the approximation (16) and the assumptions (a)–(c). This new insight suggests to us a method of improving the Langevin formalism. That is, we can go beyond the Langevin equation (38) by taking account of the second equation, i.e., the equation for $C(xt; x't')$, in the hierarchy. If we wish to take all the higher order equations in the hierarchy into account, we use Mori's Langevin-type rigorous equation⁽¹¹⁾ instead of the Heisenberg equation. Then we can obtain the most general kinetic equation. In the case of classical monatomic liquids, this approach has been studied recently by Akcasu *et al.*^(12,13) and Mazenko.⁽¹⁴⁾

It should be noted that (36) and (39) are identical in form to the Boltzmann equation for the singlet density. This fact that the singlet density and the time-correlation function obey the same kinetic equation is known in kinetic theory of classical monatomic liquids,^(12,15) and the present study confirms that this is also the case with the general nonequilibrium and nonstationary systems.

The kinetic equation (36) is one of the main results of this paper. The correlation function $F(xt; x't')$ plays a fundamental role in *nonstationary* reactor noise analyses,⁽¹⁶⁾ i.e., in detecting anomalous behavior of nuclear reactors. To calculate this correlation function, we can now make use of the methods of solution of the Boltzmann equation, which have been developed so far extensively in connection with the singlet density.

The foregoing analysis is based on the simplifying assumptions that a neutron interacts independently with each of the nuclei, that there is no neutron source, and that only one neutron is emitted per fission. These assumptions will be removed in the appendix, with the following result:

$$\begin{aligned} (\partial/\partial t)F(xt; x't') &= -B(x)F(xt; x't') \\ &\quad + S(x, t)[1 - L^3F(x, t)]F(x', t') \quad \text{for } t > t' \end{aligned} \quad (40)$$

where $B(x)$ is now defined by (A.2) and $S(x)$ represents the neutron source intensity.

4. KINETIC EQUATION FOR THE SINGLET DENSITY

Let us proceed to a kinetic equation for the singlet density. If only the direct scattering take place in the system, the nuclei can be regarded as point

particles, and the interaction potential V given by (6) is self-evident. In such a case, we generalized in II the usual transport equation so as to include the effect of multiple scattering. We can now easily extend the formalism of II to take account of nuclear reactions.

Upon neglecting the correlation, we can put $F(xt; x't') = F(x, t)F(x', t')$ and obtain from (40) the following kinetic equation for the singlet density:

$$(\partial/\partial t)F(x, t) = -B(x)F(x, t) + S(x, t)[1 - L^3F(x, t)] \quad (41)$$

In this case the approximation (16) is not necessary, and the transport equation (41) follows from the decoupling, by means of assumptions (a)–(c) in Section 3.1, of a hierarchy of equations for density functions (i.e., the BBGKY hierarchy generalized to the case of quantum mechanical systems with nuclear reactions). If the quantum effects arising from the multiple scattering and from the exclusion principle can be neglected, the transport equation (41) reduces to the usual one.

Other corrections can also be formulated in the present formalism; they are the interaction between the neutrons, the coupling of direct and resonant interactions like $T^{(W)}GT^{(V)}$, higher order interactions involved in $T^{(W)}$, such as a succession of resonance scattering and fission, and so on. These corrections, however, do not seem to be more important than the multiple scattering correction. That is, no significant correction appears even when the treatment in II is generalized to take nuclear reactions into consideration. The conclusion on the validity presented in II, therefore, applies also to the usual neutron transport equation; i.e., the validity depends on the energy of the neutron under consideration, the energy spectrum of all the neutrons, and the spatial resolution L .

5. SUMMARY AND REMARKS

In this paper we have developed a new method for finding kinetic equations in nonequilibrium systems. The kinetic equation (40) has been found, and it has been concluded that the approximation involved in the simple Langevin equation is interpreted (except for the quantum effects) as the simplest decoupling of the hierarchy of rigorous equations; an improvement of the description based on the simple Langevin equation has been suggested. The Boltzmann equation (41) for the singlet density has been derived. Finally, the quantum corrections to the usual kinetic equations have been discussed.

The present formalism is, of course, applicable to other nonequilibrium systems by suitably changing the Hamiltonian. If Girardeau's Hamiltonian⁽⁵⁾ is used, we can obtain the kinetic description of partially ionized plasmas. The Hamiltonian for other systems can easily be derived by means of the Girardeau formalism⁽⁵⁾ and of III and IV. For the case of monatomic gas, on the other hand, the present formalism yields the result of I for the singlet density and the Boltzmann-type equation of Nelkin *et al.*⁽¹⁵⁾ for the time-correlation

function. In these and other applications, the present formalism is particularly useful for dealing with higher order interactions involved in nuclear reactions, resonance scattering of electrons or photons by atoms, and so on.

The present investigation has also demonstrated the effectiveness of the Hamiltonian, like (3), for nonequilibrium systems. We therefore expect that modern statistical physics based on field-theoretical methods also is useful for discussing nonequilibrium systems. For example, we consider the two-time Green's function $\langle T\eta_{\mathbb{H}}(x_1, t_1)\eta_{\mathbb{H}}^\dagger(x_1', t_1') \rangle / i\hbar$ instead of the singlet density, and generalize the Kadanoff-Baym equations⁽¹⁷⁾ so as to include the effect of production and absorption of particles. Then, solution of the transport equation is reduced to calculation of the self-energy parts with the aid of the diagram technique. Such an approach to nonequilibrium statistical physics is now in progress.

APPENDIX

In this appendix we shall remove the three assumptions in order to derive (40).

To take account of the multiple scattering, we add the terms

$$\sum_{\alpha_U} \sum_{x', \alpha'_U} \eta^\dagger(x) \alpha_U^\dagger(x, \alpha_U | T | x', \alpha'_U) \alpha'_U \eta(x') \quad (\text{A.1})$$

to the binary-interaction approximation (26), where

$$\alpha_U^\dagger = (U_1^\dagger)^{n_1} (U_2^\dagger)^{n_2} \dots (U_\infty^\dagger)^{n_\infty} / (n_1! \dots n_\infty!)^{1/2}$$

with n_i denoting the occupation number of the i th state of nucleus U , and where the $|\alpha_U\rangle$ are occupation-number states of many U -type nuclei (e.g., two U -type nuclei in the case of double scattering). To take account of a source that emits neutrons through, e.g., photonuclear reaction $P(\gamma, n)Q$, we add to the Hamiltonian (3) the interaction potential $W(P; \gamma P) + W(P; nQ)$ and the free Hamiltonians of the nuclei P and Q . Furthermore, to take account of fission processes that emit, e.g., two neutrons because of the further decay $a \rightarrow n + b$ of the fragment a , we consider the additional Hamiltonian $H_{ob} + W(a; nb)$.

Then, it is straightforward to obtain the kinetic equation (40), with the operator $B(x)$ now given by

$$\begin{aligned} & B(x)F(xt; x't') \\ &= \frac{\mathbf{P}}{m} \cdot \nabla_{\mathbf{x}} F(xt; x't') + \frac{P}{m} \Sigma_{\text{tot}}(x)F(xt; x't') \\ &\quad - \sum_{\mathbf{P}''} \frac{P''}{m} \{ \Sigma_s(\mathbf{X}\mathbf{P}'' \rightarrow \mathbf{X}\mathbf{P}) + \bar{v}(\mathbf{P}'') \Sigma_f(\mathbf{X}\mathbf{P}'') \bar{\chi}(\mathbf{P}) \} \\ &\quad \times [1 - L^3 F(\mathbf{X}\mathbf{P}, t)] F(\mathbf{X}\mathbf{P}''t; x't') \end{aligned} \quad (\text{A.2})$$

Here, the differential and total cross sections for direct scattering are defined by (II 42) and (II 43), respectively, which include the contribution from all orders of multiple scattering occurring in the \mathbf{X} cell; $\Sigma_t(\mathbf{X}\mathbf{P}'')$ is the total fission cross section, $\bar{\chi}(\mathbf{P}'')$ is the average momentum distribution of the prompt neutrons, and $\bar{\nu}(\mathbf{P}'')$ is the average number of prompt neutrons emitted per fission. In (40), $S(x, t)$ is the source intensity given by

$$S(x, t) = \frac{1}{L^3} \sum_Q \sum_{\mathbf{q}, \lambda, P} \omega(\mathbf{q}\lambda P \rightarrow xQ) [1 \pm n(Q, t)] n(P, t) n(\mathbf{q}\lambda, t) \quad (\text{A.3})$$

where

$$\omega(\mathbf{q}\lambda P \rightarrow xQ) = \frac{2\pi}{\hbar} \delta(E(\mathbf{P}, Q) - E(\mathbf{q}, P)) |(xQ|T|\mathbf{q}\lambda P)|^2$$

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