

**THE INTERPLAY BETWEEN COHERENT
AND STOCHASTIC EFFECTS
IN DEEPLY INELASTIC COLLISIONS OF HEAVY IONS**
(I). The self-consistent mean-field approximation

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Abstract: We present a theory which describes in a consistent and uniform manner the development of the deeply inelastic collision through all its stages. We show how the two collision partners, which were initially in their ground states, become excited in the approach phase mainly through coherent processes. Gradually, denser regions of the internal excitation spectrum are populated, and a statistical description in terms of stochastic rate equations naturally emerges. The interaction between the collective degrees of freedom and the internal system is treated in a self-consistent way: The heating up of the internal system is due to the energy dissipated from the collective motion, while the force acting on the collective coordinates is derived from the internal density matrix. The internal system is discussed in the adiabatic representation. Therefore, effects which are due to the development of large deformations can be accounted for in a reliable manner. We show that our ability to describe coherent processes is intimately connected with the time resolution implicit in our quantum-statistical approach. The present formalism introduces a finer time scale to the description of DIC than has hitherto been applied, and we investigate its consequences.

1. Introduction

Much of our understanding of the deep inelastic collision (DIC) of heavy nuclei, is derived from phenomenological classical models in which only a few macroscopic degrees of freedom are treated dynamically. [See e.g. the review articles refs. ^{1–3}), and the papers cited therein.] One usually considers the relative motion of the collision partners, as well as their orientation, deformations and the mass or charge partitions as the relevant classical degrees of freedom. Classical trajectories in this

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configuration space are used to describe the time development of the reaction. To account for the large energy and angular momentum losses, phenomenological dissipative forces are incorporated in the classical equations of motion.

These models together with extensive analysis of experimental data have established time scales for DI processes. Reaction times are typically of the order of 10^{-21} s, during which the ion-ion system rotates and mass transfer occurs. Much of the energy loss occurs over a shorter time scale, around 10^{-22} s. The energy loss associated with the slower development of deformations happen over the remaining contact time.

Classical trajectory calculations are not able to account for the widths of the distributions of experimentally observed quantities. For this reason, it is recognized that fluctuations associated with the motion must be taken into account. The most straightforward way to do this is to regard the classical coordinates as being in contact with a heat bath at temperature T , in a manner analogous to the theory of the Brownian motion⁴). In the heavy-ion case, the heat bath is provided by the large number of internal nucleonic degrees of freedom. One can then derive a Fokker-Planck equation for the classical phase-space density function $\sigma(R, P, t)$. (Here R and P stand for the set of classical coordinates and their conjugate momenta, respectively.) The Fokker-Planck equation is valid only when the bath is in quasi-equilibrium, that is, when its temperature changes slowly as a result of the interaction of the bath with the collective motion. This condition is certainly not fulfilled in the early stages (the approach phase) of the deep inelastic process. Here, the two nuclei are initially in their ground states, and before close contact is established the nuclear temperature is raised to 1–2 MeV within a very short time interval. Once this temperature is reached, quasi-equilibrium prevails and transport theories may be justified.

To date, most theoretical efforts have been directed towards the investigation of the quasi-equilibrium stage (the contact phase) in DIC^{5–7}). Appropriate transport equations have been derived and the relevant transport coefficients determined, from “basic” principles and nuclear properties. No satisfactory theory has been put forward in which both the approach and the contact phases can be described, even though the need for such a development has repeatedly been stressed^{8–10}). The purpose of this work is to develop a formalism which provides a uniform description of the DIC process, uniform in the sense that the approach and contact phases, as well as the transition between them, are accounted for within the same theory. This theory also treats consistently the intrinsic and collective dynamics, and takes full account of the correlations between the two sub-systems. That is, the fluctuations in the collective variables and hence the width of their final distributions are completely accounted for. Such a theory is necessary in order to describe the smooth transition between deeply inelastic and “quasi-elastic” collisions, and to investigate the role of entrance channel characteristics in determining the outcome of such reactions.

We shall present our theory in the following way: Once the “collective” and the intrinsic degrees of freedom are defined, one may describe the development of the entire system using the semi-classical Liouville equation ⁴⁾

$$\frac{d}{dt} \hat{D}(R, P, t) = \frac{1}{i\hbar} [\hat{H}(R, P), \hat{D}(R, P, t)]_- - \frac{P}{M} \frac{\partial}{\partial R} \hat{D}(R, P, t) - \frac{\partial}{\partial P} [\hat{F}, \hat{D}(R, P, t)]_+. \quad (1.1)$$

The operators appearing as \hat{O} in (1.1) are matrices in the intrinsic subspace and functions of the classical collective phase-space variables R, P . They are obtained by a partial Wigner transform, taken with respect to the collective degrees of freedom only. $\hat{D}(R, P)$ is the density operator, so that

$$\sigma(R, P, t) = \text{tr } \hat{D}(R, P, t) \quad (1.2)$$

is the collective phase-space density function, and

$$\hat{\rho}(t) = \frac{1}{2\pi\hbar} \int dR \, dP \, \hat{D}(R, P, t) \quad (1.3)$$

is the internal density matrix.

The other operators in (1.1) are the hamiltonian \hat{H} and the force operator $\hat{F} = -\partial\hat{H}/\partial R$. $[,]_-$ stand for the commutator and the anti-commutator, respectively.

Before the collision, when $t \rightarrow -\infty$, $\hat{D}(R, P, t)$ describes the two collision partners moving towards each other with relative velocity P_0/M , and the internal system in its ground state.

$$\hat{D}(R, P, t) \xrightarrow[t \rightarrow -\infty]{} \delta\left(R - \frac{P}{M}t\right) \delta(P - P_0) |0\rangle\langle 0|. \quad (1.4)$$

Subject to the initial condition (1.4), eq. (1.1) governs the development of the reaction. In the present work we propose a scheme of approximations which will make the solution of (1.1) tractable, and retain the interesting physics.

To facilitate the presentation, we report our results in two publications in which solutions of the above-mentioned problem will be given, differing in the extent to which the correlations between the collective and the intrinsic fluctuations are considered.

The major assumption in the present paper (I) is that the density operator \hat{D} can be factored at all times:

$$\hat{D}(R, P, t) = \hat{\rho}(t) \sigma(R, P, t). \quad (1.5)$$

Substituting (1.5) into (1.1) one gets

$$\frac{d}{dt} \sigma(R, P, t) = -\frac{P}{M} \frac{\partial \sigma}{\partial R} - \langle \hat{F} \rangle_\rho \frac{\partial \sigma}{\partial P}, \quad (1.6)$$

$$\frac{d}{dt} \hat{\rho}(t) = \frac{1}{i\hbar} [\langle \hat{H} \rangle_\sigma, \hat{\rho}]_-, \quad (1.7)$$

with

$$\langle \hat{F} \rangle_\rho = \text{tr} [\hat{F} \rho], \quad (1.8)$$

$$\langle \hat{H} \rangle_\sigma = \frac{1}{2\pi\hbar} \int dP dR \sigma(R, P, t) \hat{H}(R, P, t). \quad (1.9)$$

Eq. (1.6) allows a description of the collective motion in terms of classical trajectories. Writing

$$\sigma(R, P, t) = \delta(R - R(t))\delta(P - P(t)) \quad (1.10)$$

and substituting in (1.6) we get

$$\dot{R} = P(t)/M, \quad (1.11)$$

$$\dot{P} = \langle \hat{F}(R) \rangle_\rho, \quad (1.12)$$

while (1.7) is reduced to

$$\frac{d}{dt} \hat{\rho} = \frac{1}{i\hbar} [\hat{H}(R(t), P(t)), \hat{\rho}]_- . \quad (1.13)$$

Eqs. (1.11)–(1.13) are the starting points for the theory presented below. Due to the ansatz (1.5) the correlations between the collective and the intrinsic degrees of freedom are neglected. The two subsystems affect each other *in the mean*, and self-consistently. This approximation is known in the literature as the self-consistent mean field approximation^{11–12)} (SCMFA). Even though eqs. (1.11)–(1.13) are much simpler than the original eq. (1.1), it is impossible to solve them exactly because of the huge dimensionality of the internal space. We therefore must develop a reduced description, in which only coarse-grained internal variables are considered. Because of the time scales involved in our system, and the initial condition (1.4), we are obliged to consider very carefully the choice of variables which will be included in the reduced equations of motion. This is the major issue common to the two approaches to be described in this series of papers. It will be discussed in greater detail in the present paper, since here it appears in a more transparent context.

In the second paper in the series we shall amend the most serious drawback of paper I, namely, we shall discuss the fluctuations in the collective motion which are due to the correlations between the intrinsic and collective subsystems. This is done by writing the equations of motion for $\sigma(R, P, t)$ and $\hat{\rho}(t)$ as defined by eq. (1.2)–(1.3) without invoking the separability ansatz (1.5). One gets for $\sigma(R, P, t)$ a generalized Fokker–Planck equation, coupled to a von Neumann equation of the type (1.7). In the conventional application of the Fokker–Planck equation for quasi-equilibrium situations, one *assumes*⁴⁾ a canonical distribution for the internal

system, i.e. $\hat{\rho}(t) \propto \exp(-\langle \hat{H} \rangle_{\sigma}/T)$. Using this assumption, and recognizing the difference in the time scales which characterize the two sub-systems, one can extract the transport coefficients which appear in the Fokker-Planck equation. The theory presented in paper II of this series describes a different situation, in which the intrinsic system is not in quasi-equilibrium but develops with time so that the effective transport coefficients for the generalized Fokker-Planck equation are deduced at each moment from the statistical matrix $\hat{\rho}(t)$. This approach does not take into account all correlations between the collective and the internal sub-systems. The statistical matrix $\hat{\rho}(t)$ is determined by using information from the *mean* collective trajectory. Thus, the fluctuations which are induced in the internal motion because of fluctuations in the collective sub-system are neglected. In the second part of paper II we shall present a theory which corrects this deficiency. The starting point for the approximation scheme is now equation (1.1), and the coarse-graining is imposed on $\hat{D}(R, P, t)$ itself. One obtains a complicated set of coupled equations for the coarse-grained dynamical variables, which is then used to derive new approximate, reduced equations of motion.

It should be stressed once again that the common feature of the three approaches is that the internal system is not assumed to be in a quasi-equilibrium state, but that it develops from its cold initial state as the collision progresses in time.

As mentioned before, this paper presents results obtained within the self-consistent mean-field approximation, eqs. (1.11)-(1.13). In sect. 2 we define the internal system and formulate the equations of motion in the adiabatic basis. The coarse-graining procedure and the choice of dynamical variables are discussed here too. In sect. 3 projection operator techniques are used to derive reduced equations of motion (REM) for expectation values of the dynamical variables. Subsequent analysis of the equations of motion shows that they indeed provide a uniform description of the transition from the coherent to the statistical phase in DIC. The expression of the induced force (1.8) in terms of expectation values of the dynamical variables which describe the internal system is derived in sect. 4. This expression is compared to the dissipative (friction) force obtained from perturbation theory. It is shown that the general expression obtained here reduces to the perturbative result in the corresponding limit. We conclude this paper with a short summary.

2. The model space and the adiabatic representation

As discussed in the introduction, we describe DIC in terms of a few collective degrees of freedom (the separation between the centers of mass of the reactants, their deformations and orientation in space) and a large number of internal degrees of freedom, which account for the internal states of the colliding nuclei. There are two basic problems connected with this classification. A rigorous definition of collective variables must be performed through a canonical transformation of the $3A$ nucleonic degrees of freedom, to K collective variables plus $3A - K$ internal

coordinates. Such a procedure was investigated by e.g. Agassi¹³⁾, and it introduces almost prohibitive complications to the resulting equations of motion. We shall therefore adopt the conventional simplification and impose the collective variables in a phenomenological way. This simplification is justified when $3A \gg K$. The second problem is the fact that our choice of collective variables depends on the particular transfer channel one considers. Again we shall adopt the simplest approach, and neglect the effects of mass transfer on the definition of the collective coordinates and the associated mass tensors. This is justified because the mean mass transfer in most DIC is rather small, and because the collisions are quite peripheral.

When the collision partners are well separated, one can define an asymptotic basis, in terms of which the internal excitations will be described. We denote the asymptotic internal states by $|n\nu\rangle$, where n refers to a particular mass and charge partition (channel) between the two fragments, and ν stands for all the quantum numbers which specify the internal state for a given n . We let $n = 0$ be the entrance channel partition.

In preliminary reports of our work¹⁴⁾ we used the asymptotic vectors discussed above as a basis for the description of the entire process (the *diabatic* representation). This basis has a few advantages, especially when one is studying early stages of the collision, before large deformations set in. Here, we shall rather use the *adiabatic* representation, since it provides a better starting point for approximations which are necessary in the description of the contact phase.

The adiabatic basis is constructed in the following way: The equations of motion (1.11)–(1.13) read,

$$i\hbar\dot{\rho} = [\hat{H}_0 + \hat{V}(R(t)), \hat{\rho}], \quad (2.1)$$

$$\dot{R} = \dot{P}/M, \quad \dot{P} = \langle \hat{F}(R) \rangle = -\text{tr}[\hat{\rho}\nabla\hat{V}], \quad (2.2)$$

where \hat{H}_0 is independent of the collective variables R and determines the internal structure of the two non-interacting nuclei. Thus, \hat{H}_0 is built of one- and two-body operators, while \hat{V} , which describes the influence of one nucleus on the nucleons in the other fragment, is primarily a one-body operator. \hat{V} depends on time through its arguments $R(t)$. For any time t , we define the adiabatic basis as the set of eigenstates of the operator $\hat{H}_0 + \hat{V}(R(t))$, which satisfy

$$[\hat{H}_0 + \hat{V}(t)]|n\nu\rangle_t = E_{n\nu}(t)|n\nu\rangle_t, \quad (2.3)$$

$$|n\nu\rangle_t \xrightarrow[t \rightarrow -\infty]{} |n\nu\rangle. \quad (2.4)$$

That is, we use the asymptotic (diabatic) basis to *label* the adiabatic states at each time. We use round brackets to denote the diabatic basis, as indicated by eq. (2.4). We shall omit the time indicator. The phase of the states $|n\nu\rangle$ is not specified by the requirements (2.3)–(2.4). We choose it in such a way that the following

condition is satisfied:

$$\frac{d}{dt}|m\mu\rangle = \sum'_{n\nu} \frac{\langle n\nu | \hat{V} | m\mu \rangle}{E_{m\mu} - E_{n\nu}} |n\nu\rangle. \quad (2.5)$$

The primed summation symbol means $m\mu \neq n\nu$, and

$$\hat{V} = \dot{R} \cdot \nabla \hat{V}. \quad (2.6)$$

In the adiabatic (moving) frame, eq. (2.1) reduces to

$$i\dot{\rho}_{n\nu,m\mu} = \frac{1}{\hbar}(E_{n\nu} - E_{m\mu})\rho_{n\nu,m\mu} + [\hat{v}, \hat{\rho}]_{n\nu,m\mu}, \quad (2.7)$$

where

$$\dot{\rho}_{n\nu,m\mu} = \begin{cases} \frac{i\langle n\nu | \hat{V} | m\mu \rangle}{E_{n\nu} - E_{m\mu}}, & n\nu \neq m\mu \\ 0, & n\nu = m\mu. \end{cases} \quad (2.8)$$

Here $\dot{\rho}_{n\nu,m\mu}$ means $d\langle n\nu | \rho | m\mu \rangle / dt$. One must remember that in the adiabatic basis the time derivative of a matrix element is not the matrix element of the time derivative. It simplifies the analysis if one uses only matrices in the basis, and not Hilbert space operators.

Because of the large number of states in the internal system, and the complicated structure of the individual coupling matrix elements, it is impossible to obtain a detailed solution of eqs. (2.1) and (2.2). We can get a manageable system of equations and satisfactory description of the collision by developing a coarse-grained theory. For this purpose, the spectrum of each mass-charge partition, n , is subdivided into excitation energy bins. These bins are chosen to be sufficiently large so that they contain very many states, and statistical arguments can be invoked within each bin. At the same time they are sufficiently small (1 or a few MeV, for example) so that we have no interest in the probability distribution *within* a bin: the bin is smaller than the relevant experimental energy resolution. It is easy to choose bins compatible with these two conditions because DIC deal with very large excitation energies (≥ 100 MeV), and so the density of levels – although not well known – is certainly extremely large ($\gg 1$ MeV⁻¹).

It should be emphasized that the coarse-graining is defined for the internal spectrum of the separated nuclei. Once the nuclei come into close contact the order of levels may change and bins may overlap. Indeed, bins for different mass-charge partitions will overlap even before and after the contact phase. As we shall see below, there will be transfer of probability between bins only when the energy difference between bins is not much greater than the interaction matrix element.

In what follows, the single index n will refer to both the mass-charge value and the excitation energy bin.

While we are primarily interested in the dense statistical levels at high energies for describing DIC, there may be a small number of states at low excitation which play a special role, especially during the initial stages of the collision. These states will be labeled with a single upper case index, $|I\rangle$. The ground state in the $n = 0$ partition must certainly be in this set, and we call it $|0\rangle$. The ground state in other partitions, and a few isolated low-lying states in $n = 0$ (or in other partitions) may have to be treated separately in the approach phase.

Note, however, that if deformation coordinates are included among the collective degrees of freedom then it would not be appropriate to treat low-lying vibrational levels as special states, since that would be accounting for the same phenomenon twice. We must use some insight in the choice of variables and states. Similarly for rotational motion.

There may be some interest in investigating the role of giant multipole resonances in the approach phase – or the contact phase. Since these are collections of large numbers of states, of a given spin and parity, each giant resonance may be thought of as a single bin.

The operators which introduce the coarse graining are the Hilbert space projectors for each bin, $\hat{a}(n)$, which have matrix elements

$$a_{l\lambda,m\mu}(n) = \frac{1}{d_n^{1/2}} \delta_{ln} \delta_{mn} \delta_{\lambda\mu}, \quad (2.10)$$

where d_n is the number of states in this bin. The population probabilities of the bins are related to the expectation value of the $a(n)$ through

$$P_n(t) = d_n^{1/2} \text{tr} [a(n)\hat{\rho}] = \sum_{\nu} \rho_{n\nu,n\nu}. \quad (2.11)$$

In an analogous manner we define

$$a_{JK}(I) = \delta_{IJ} \delta_{IK} \quad (2.12)$$

(all other matrix elements being zero), which relate to the special states discussed above.

In the conventional treatment of pre-equilibrium statistical mechanics ¹⁵⁾, one assumes that the expectation values of the population matrices $a(n)$ provide sufficient information about the collision process. One also neglects the $a(I)$ matrices since the internal system is assumed to be initially in a complicated non-coherent state. One obtains a set of generalized master equations for the $P_n(t)$ which read

$$\frac{dP_n}{dt} = \sum_m 2\gamma_{nm}^2(t) \int_{-\infty}^t dt' \text{Re} [\tilde{I}_{nm}(t, t')] [P_m(t')/d_m - P_n(t')/d_n]. \quad (2.13)$$

Here, $\gamma_{nm}(t)$ measures the coupling between the m, n bins,

$$\gamma_{nm}^2(t) = \sum_{\nu\mu} |v_{n\nu,m\mu}(t)|^2 \equiv (d_n d_m)^{1/2} \Omega_{nm}^2, \quad (2.14)$$

and \tilde{I}_{nm} is an internal correlation function which, to lowest order in v , becomes

$$\tilde{I}_{nm}(t, t') = \frac{1}{\gamma_{nm}^2(t)} \sum_{\nu\mu} v_{n\nu, m\mu}^*(t) v_{n\nu, m\mu}(t') \exp \left[-(i/\hbar) \int_{t'}^t (E_{n\nu}(t'') - E_{m\mu}(t'')) dt'' \right]. \quad (2.15)$$

$\tilde{I}_{nm}(t, t')$ is normalized to unity at $t = t'$. As $t - t'$ increases the various oscillating terms in eq. (2.15) become out of phase, and \tilde{I} decreases. For relatively long time differences one expects it will decrease with a characteristic time constant τ^d (dephasing time). Experience¹⁶⁾ with statistical theories in general indicates that correlation functions behave in the manner shown in fig. 1, where there is a correlation time, τ^c , before the characteristic decay begins. The correlation function in fig. 1 is taken from the theory of Brownian motion. We chose a non-realistic case where $\tau^c \sim \tau^d$ only to demonstrate the behaviour of \tilde{I} in the two regimes.

The validity of any statistical theory requires that one assumes $\tau^c \ll \tau^d$, and this assumption is the basis of what follows in the present paper. At the same time, one assumes that $\tau_{nm}^d \ll \Omega_{nm}^{-1}$, in order to invoke the Markovian approximation,

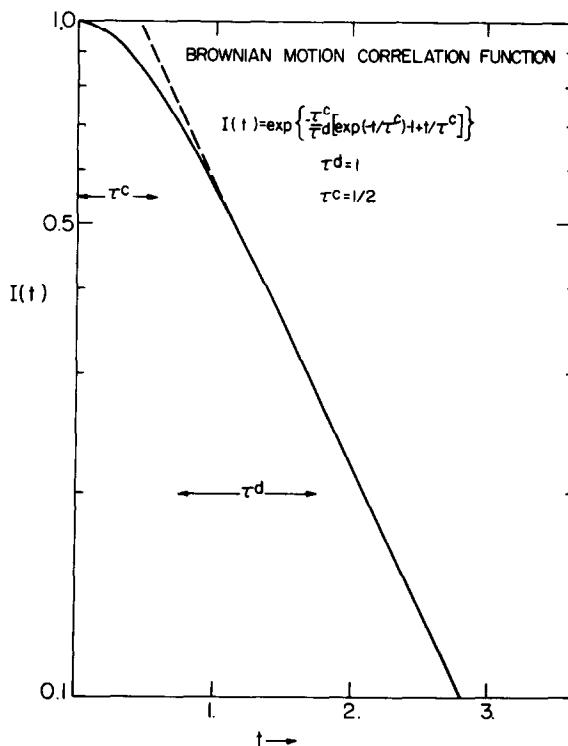


Fig. 1. The two-time correlation function $I(t, 0)$ taken from the theory of Brownian motion.

which amounts to replacing $\tilde{I}_{nm}(t, t')$ by $\tau_{nm}^d \delta(t - t')$. One then obtains the conventional time-local master equation

$$\frac{dP_n}{dt} = 2 \sum_m \gamma_{nm}^2(t) \tau_{nm}^d (P_m(t)/d_m - P_n(t)/d_n). \quad (2.16)$$

We stress that the loss of memory achieved here, resulting from the short time τ^d , is directly connected to the strength of the interaction, Ω_{nm} and v_{nm} [eq. (2.14)]. The condition $\Omega_{nm}^{-1} \gg \tau_{nm}^d$ implies that the rate of change of the populations, $\Omega_{nm}^2 \tau_{nm}^d$, is much slower than the decay (dephasing) time τ_{nm}^d . By adopting the Markovian approximation at this point, we give up the ability to resolve variations at the time scale of τ_{nm}^d . As long as one is interested in describing the contact phase in DIC, this time resolution is sufficient. A better resolution is needed to describe the fast processes which occur during the approach phase. In developing a theory for these processes, one may not use equations of the type (2.16) and must choose between two alternative courses. One can either retain the form (2.13) and solve an integro-differential set of equations or enlarge the set of dynamical variables and include those variables which vary on the time scale of τ^d . When taking this alternative, one may obtain time local equations of motion by imposing a Markovian approximation on a *finer* level of time resolution than was implied in obtaining (2.16). When contemplating the latter alternative, one should always remember that memory effects are due to the amount of information one must sacrifice in the transition from the original von Neumann equation to the reduced description. When no reduction is imposed, one retains complete information about the system and the equations of motion do not contain any memory effects.

In the present work we increase the set of dynamical variables by following the procedure proposed recently for molecular multiphoton processes¹⁸⁾. This scheme is based on the construction of an appropriate Mori hierarchy^{17,19)} of dynamical variables whose expectation values change on time scales which become progressively shorter. Starting with the population matrices, $a(n)$, we generate their time derivatives $[v, a(n)]$ as linear combinations of the matrices

$$A_{l\lambda, k\kappa}(n, m) = \gamma_{nm}^{-1} \delta_{ln} \delta_{km} v_{l\lambda, k\kappa}, \quad n \neq m, \quad (2.17)$$

with

$$\gamma_{nm}^2 = \sum_{\nu\mu} |v_{n\nu, m\mu}|^2. \quad (2.18)$$

The normalization coefficients γ_{nm} are introduced for a purpose which will become clear later.

The matrices $A(n, m)$ will be added to the $a(n)$ to form the extended set of dynamical variables whose expectation values describe the development of the system. The expectation values of $A(n, m)$ define new variables S_{nm} (coherences) through the relation

$$S_{nm}(t) = \gamma_{nm}(t) \operatorname{tr} [A^+(n, m)\rho] = \sum_{\nu\mu} v_{n\nu, m\mu}^* \rho_{n\nu, m\mu}. \quad (2.19)$$

Since the S_{nm} are related to off-diagonal elements of the density matrix, they describe coherent (i.e. quantal) effects in the time development of the system (even though averages over bins are taken).

The equations of motion to be developed in this work will be written in terms of the $P_n(t)$ and $S_{nm}(t)$. Once these equations are given it will become clear that the temporal dependence of the $S_{nm}(t)$ is determined by the properties of the function $\tilde{I}_{nm}(t, t')$ defined above, so that the time resolution gained by enlarging the set of operators is sufficient to describe variations on the scale of τ_{nm}^d . For the present application this is sufficient and we shall not add any further dynamical variables. One could do so by considering the operators which are needed to construct the higher derivatives of $a(n)$, namely, $[v, [v, a(n)]]$, etc. ^{18,20}). In a similar fashion to (2.17) one also defines coherences related to the special states $|I\rangle$. Thus

$$A_{KL}(I, J) = \delta_{IK}\delta_{JL}, \quad (2.20)$$

$$A_{m\mu,J}(n, I) = \gamma_{nI}^{-1} \delta_{nm} \delta_{IJ} v_{m\mu,J}, \quad (2.21)$$

with

$$\gamma_{nI}^2 = \sum_\nu |v_{n\nu,I}|^2, \quad (2.22)$$

and all other matrix elements vanish.

Considering $A(I, J)$ and $a(I)$, we see that the space spanned by these matrices is the *entire* $|I\rangle$ subspace. Hence, the reduced equations of motion which we shall obtain will give *complete* information about this subspace. The matrices $A(n, I)$ are responsible for transitions from the special subspace to the statistical coarse-grained states.

In summary, our need to describe the collision during the initial stage of DIC, forces us to discuss it on a finer time scale than the one required to describe the contact phase, and to recognize the special role of the ground state $|0\rangle$ and the states $|I\rangle$ which are strongly coupled to it. In order to do so we have to extend the set of dynamical variables to include operators which project on the non-diagonal elements of the density matrix. The implicit assumption that this set exhausts the entire space of slow and relevant variables will be discussed in a future publication.

Up to this point the discussion has concentrated on the treatment of eq. (2.1). When we consider its counterpart (2.2), we see that at each moment of time one must supply information about the induced forces, $\langle \hat{F}(R) \rangle$. The dissipative part of the force is intimately related to the off-diagonal elements of the density matrix, and therefore we cannot calculate it in terms of its projection on the set $a(n)$ alone. In sect. 4 we shall show how $\langle \hat{F}(R) \rangle$ can be related to the coherences S_{nm} .

3. The reduced equations of motion

In the present section we shall derive the reduced equations of motion (REM), for the populations and coherence operators which were defined in the preceding

section. It is convenient to rewrite the von Neumann equation (2.7) as

$$i|\dot{\rho}\rangle\langle = L|\rho\rangle\langle, \quad (3.1)$$

where we consider the density matrix $\hat{\rho}$ as a tetricadic vector¹⁵⁾ and the liouvillian L as a tetricadic matrix.

$$\begin{aligned} L &= L^{(0)} + L^{(1)}, \\ L_{l\lambda, k\kappa; n\nu, m\mu}^{(0)} &= (E_{l\lambda} - E_{k\kappa})\delta_{l\lambda, n\nu}\delta_{k\kappa, m\mu}, \\ L_{l\lambda, k\kappa; n\nu, m\mu}^{(1)} &= v_{l\lambda, n\nu}\delta_{m\mu, k\kappa} - \delta_{l\lambda, n\nu}v_{m\mu, k\kappa}. \end{aligned} \quad (3.2)$$

Also,

$$(L\rho)_{l\lambda, k\kappa} = \sum_{m\mu, n\nu} L_{l\lambda, k\kappa; n\nu, m\mu} \rho_{n\nu, m\mu}. \quad (3.3)$$

An inner product of two tetricadic vectors is defined by

$$\langle A | B \rangle = \text{tr}(A^+ B). \quad (3.4)$$

Using this notation the populations and coherences defined in the preceding section can be written as

$$P_n = \langle a(n) | \rho \rangle d_n^{1/2}, \quad (3.5)$$

$$S_{nm} = \langle A(n, m) | \rho \rangle \gamma_{nm}. \quad (3.6)$$

The combined set of tetricadic vectors which corresponds to the coherence and population operators is an orthonormal set,

$$\begin{aligned} \langle a(n) | A(k, l) \rangle &= 0, \\ \langle A(k, l) | A(m, n) \rangle &= \delta_{km}\delta_{ln}, \\ \langle a(n) | a(m) \rangle &= \delta_{nm}. \end{aligned} \quad (3.7)$$

It spans the subspace of operators which are relevant to our coarse-grained description of the collision process. We project onto this subspace by the tetricadic projector

$$\pi = \sum_n |a(n)\rangle\langle a(n)| + \sum_{kl} |A(k, l)\rangle\langle A(k, l)|, \quad (3.8)$$

or, explicitly

$$\pi_{l\lambda, k\kappa; n\nu, m\mu} = d_l^{-1}\delta_{nl}\delta_{n\nu, m\mu}\delta_{l\lambda, k\kappa} + \gamma_{lk}^{-2}\delta_{nl}\delta_{mk}v_{n\nu, m\mu}^*v_{l\lambda, k\kappa}. \quad (3.9)$$

Although we have not made explicit reference to the operators $a(I)$, $A(I, J)$ and $A(I, n)$, they are included in all the relevant definitions and summations. One can think of the states $|I\rangle$ as belonging to “bins” which contain only one state, with $\gamma_{II} = d_I = 1$, while γ_{In} was defined previously in eq. (2.22).

Another important point is that the projection operator defined above is constructed of two distinct parts. The first, which projects onto the population operators, is time independent, while the second part depends on time.

We shall now return to the von Neumann equation (3.1) and solve it subject to a slightly more general initial condition than (1.4). Let the density matrix at $t = t_0$ be $\hat{\rho}_0$, such that

$$\pi|\rho_0\rangle\langle\rho_0| = |\rho_0\rangle\langle\rho_0|. \quad (3.10)$$

The evolution operator $U(t, t')$ satisfies

$$i\dot{U} = LU, \quad U(t, t) = 1, \quad (3.11)$$

$$|\rho(t)\rangle\langle\rho(t)| = U(t, t_0)|\rho_0\rangle\langle\rho_0|. \quad (3.12)$$

Since the tetradic vector $|\rho_0\rangle\langle\rho_0|$ is in the subspace onto which π projects,

$$\pi|\rho(t)\rangle\langle\rho(t)| = (\pi U \pi)|\pi\rho_0\rangle\langle\pi\rho_0|, \quad (3.13)$$

but,

$$i|\dot{\rho}\rangle\langle\dot{\rho}| = L|\rho\rangle\langle\rho| = LU|\rho_0\rangle\langle\rho_0| = LU(\pi U \pi)^{-1}|\pi\rho_0\rangle\langle\pi\rho_0|, \quad (3.14)$$

so that

$$i\pi|\dot{\rho}\rangle\langle\dot{\rho}| = i\frac{d}{dt}(\pi|\rho\rangle\langle\rho|) - i\dot{\pi}|\rho\rangle\langle\rho| = (\pi LU \pi)(\pi U \pi)^{-1}|\pi\rho\rangle\langle\pi\rho|. \quad (3.15)$$

If we denote by $\{|M_\alpha\rangle\}$ the set of tetradic vectors $\{|a(n)\rangle, |A(k, l)\rangle\}$, we get

$$i\frac{d}{dt}\langle M_\alpha|\rho\rangle\langle\rho|M_\beta\rangle = \sum_{\beta\gamma} \langle M_\alpha|LU|M_\beta\rangle\langle M_\beta|(\pi U \pi)^{-1}|M_\gamma\rangle\langle M_\gamma|\rho\rangle + i\left(\frac{d}{dt}\langle M_\alpha|\right)|\rho\rangle. \quad (3.16)$$

Eq. (3.16) forms the desired reduced equations of motion for the expectation values $\langle M_\alpha|\rho\rangle$. The last term in (3.16) projects out of the space of relevant operators. We shall show below how it can be approximately expressed in terms of the variables $\langle M_\alpha|\rho\rangle$.

Reduced equations of motion of the type (3.16) were introduced recently for the study of multi-photon absorption by complicated molecules¹⁸). There are two important aspects in which they differ from the more conventional form of reduced descriptions. (a) They do not contain a convolution in time. (b) They involve the calculation of the propagator U of the *entire* system, rather than its counterpart which relates to the evolution of the *non-relevant space* projected by $(1 - \pi)$. A detailed discussion of this type of reduced equations of motion, and its relation to the more conventional treatment can be found in ref.²⁰).

Eq. (3.15) can also be cast in the form

$$i\pi|\dot{\rho}\rangle\langle\dot{\rho}| = (\pi L \pi)\pi|\rho\rangle\langle\rho| + \pi L(1 - \pi)U\pi(\pi U \pi)^{-1}\pi|\rho\rangle\langle\rho|. \quad (3.15')$$

Here the term which is due to the mean coupling, $\pi L \pi$, and that is acting within

the projected space, is separated from that which comes from the fluctuations (the second term). In deriving the equations for P_n and S_{nm} we shall use (3.15) directly and keep only first order terms in $L^{(1)}$. But, we observe from (3.15') that the non-fluctuating part is in fact treated exactly. Furthermore, with regard to the special states, $|I\rangle$, the projector exhausts all the operators $|I\rangle\langle J|$, and so, within the Hilbert space spanned by these states, $(1 - \pi) = 0$; the equations of motion are exact.

We shall derive approximate expressions for the coefficient matrix in (3.16) using the following reasoning: As was discussed in the preceding chapter, we require a distinct separation between the time scales which characterize the variables $S_{nm}(t)$, $P_n(t)$ and the correlation time τ^c , i.e. $\tau^d \gg \tau^c$. In other words, we tacitly assume that all the dynamical variables which change on the time scale of interest are included in the π space, while those which belong to the complementary operator space $(1 - \pi)$ vary on a time scale of the order τ^c . Let Δ be a time interval intermediate between τ^c and τ^d . If we introduce a smoothing over the time with a time interval Δ (coarse graining), any operator in the complementary space $(1 - \pi)$ would have sufficient time to decay (dephase) and therefore $\bar{U}(t_0 + \Delta, t_0)\pi \approx \pi\bar{U}(t_0 + \Delta, t_0)\pi$. Thus, within such a coarse-grained description[†]

$$\begin{aligned} & (\pi L \bar{U}(t, t_0)\pi)(\pi \bar{U}(t, t_0)\pi)^{-1} \\ & \quad \approx (\pi L \bar{U}(t, t - \Delta)\pi)(\pi \bar{U}(t - \Delta, t_0)\pi)[(\pi \bar{U}(t, t - \Delta)\pi)(\pi \bar{U}(t - \Delta, t_0)\pi)]^{-1} \\ & \quad = (\pi L \bar{U}(t, t - \Delta)\pi)(\pi \bar{U}(t, t - \Delta)\pi)^{-1}. \end{aligned} \quad (3.17)$$

We see that because of the fast decay of the operators in the $(1 - \pi)$ space, the coefficient matrix in eq. (3.16) can be approximated by its mean value over the interval Δ which precedes the actual time where they are required. We shall further assume that $\langle v \rangle \Delta \ll 1$, where $\langle v \rangle$ is a measure of the size of the matrix elements (2.8). This assumption underlies the adiabatic approximation, and guarantees that the approximation of U to this order is sufficient for time intervals which are of order Δ .

The leading terms in the adiabatic expansion of U are

$$\begin{aligned} U(t, t') &= U_0(t, t') + U_1(t, t'), \\ U_0(t, t') &= \exp \left(-i \int_{t'}^t L^{(0)}(t'') dt'' \right), \\ U_1(t, t') &= -i \int_{t'}^t U_0(t, t'') L^{(1)}(t'') U_0(t'', t') dt''. \end{aligned} \quad (3.18)$$

We expect the matrix elements $\langle nv | \partial V / \partial R | \lambda \rangle$ to behave randomly in phase as a function of λ . Similarly, a product of two matrix elements $\langle m\mu | \partial V / \partial R | \lambda \rangle$

[†] A more quantitative discussion of the coarse graining in time and a rigorous justification of eq. (3.17) is deferred to paper II of this series.

$\langle l\lambda | \partial V/\partial R | n\nu \rangle$ will have a random phase as λ is varied, provided that $m\mu \neq n\nu$. We further assume that within given bins m, l , the phase of $\langle m\mu | \partial V/\partial R | l\lambda \rangle$ is not correlated with $(E_{l\lambda} - E_{m\mu})$, so that the matrix elements $v_{n\nu, l\lambda}$ are also random in phase with respect to λ [ref. 21)].

Consequently,

$$\sum_{\lambda} v_{n\nu, l\lambda} v_{l\lambda, m\mu} \propto \delta_{n\nu, m\mu}, \quad (3.19a)$$

$$\sum_{\lambda\mu} v_{n\nu, l\lambda} v_{l\lambda, m\mu} v_{m\mu, n\nu} = 0, \quad (3.19b)$$

where the last relation is due to (3.19a) and the fact that the diagonal elements of v (2.8) vanish.

Using the expansion (3.18) and (3.19), we can explicitly calculate the coefficients in the REM (3.16). After some algebra we obtain equations for the variables P_l and S_{kl} . The relation between the time derivatives of the populations and the coherences is an *identity* which follows from our definition of these quantities, as explained in sect. 2:

$$\dot{P}_l = 2 \operatorname{Im} \sum_k S_{kl}. \quad (3.20)$$

The complementary equations for the coherences read,

$$i\dot{S}_{kl} = i\left(\frac{\overset{(.)}{I}_{kl}}{\overset{(.)}{I}_{kl}}\right)_{\Delta} S_{kl} + \gamma_{kl}^2 \left(1 + \frac{\overset{(.)}{J}_{kl}}{\overset{(.)}{I}_{kl}}\right)_{\Delta} (P_l/d_l - P_k/d_k) + if_{kl}. \quad (3.21)$$

The r.h.s. of eq. (3.21) is “diagonal” in the sense that the only indices referred to are (k, l) which appear also on the l.h.s. Off-diagonal terms of the form $\sum \chi_{kj} S_{jl}$ or $\sum S_{kj} \chi_{jl}$ vanish due to (3.19).

The coefficients in eq. (3.21) have to be understood in the spirit of the time coarse-graining which was introduced in the discussion which follows eq. (3.22). The subscript Δ indicates an average over the time interval $(t - \Delta, t)$.

The correlation functions $I_{kl}(t, t')$, $\overset{(.)}{I}_{kl}(t, t')$, $J_{kl}(t, t')$ and $\overset{(.)}{J}_{kl}(t, t')$ are given by

$$I_{kl}(t, t') = \gamma_{kl}(t)^{-2} \sum_{\lambda\kappa} |v_{k\kappa, l\lambda}(t)|^2 \exp\left[-\frac{i}{\hbar} \int_{t'}^t d\tau (E_{k\kappa}(\tau) - E_{l\lambda}(\tau))\right], \quad (3.22)$$

$$\begin{aligned} \overset{(.)}{I}_{kl}(t, t') &= -i\gamma_{kl}(t)^{-2} \sum_{\lambda\kappa} |v_{k\kappa, l\lambda}(t)|^2 (E_{k\kappa}(t) - E_{l\lambda}(t)) \\ &\times \frac{1}{\hbar} \exp\left[-\frac{i}{\hbar} \int_{t'}^t d\tau (E_{k\kappa}(\tau) - E_{l\lambda}(\tau))\right], \end{aligned} \quad (3.23)$$

$$\begin{aligned} J_{kl}(t, t') &= \gamma_{kl}(t)^{-2} \int_{t'}^t d\tau v_{k\kappa, l\lambda}^*(t) v_{k\kappa, l\lambda}(\tau) \\ &\times \exp\left[-\frac{i}{\hbar} \int_{\tau}^t d\tau' (E_{k\kappa}(\tau') - E_{l\lambda}(\tau'))\right], \end{aligned} \quad (3.24)$$

$$\begin{aligned} \overset{(1)}{J}_{kl}(t, t') = & -i\gamma_{kl}(t)^{-2} \int_{t'}^t d\tau v_{k\kappa, l\lambda}^*(\tau) v_{k\kappa, l\lambda}(\tau) \\ & \times (E_{k\kappa}(t) - E_{l\lambda}(t)) \frac{1}{\hbar} \exp \left[-\frac{i}{\hbar} \int_{\tau}^t d\tau' (E_{k\kappa}(\tau') - E_{l\lambda}(\tau')) \right]. \end{aligned} \quad (3.25)$$

These functions are closely related to each other and to the correlation function $\tilde{I}_{kl}(t, t')$ defined in eq. (2.15). We shall discuss these shortly. The quantities f_{kl} are given by

$$f_{kl} = \sum_{\kappa\lambda} \left(\frac{d}{dt} v_{k\kappa, l\lambda}^* \right) \rho_{k\kappa, l\lambda}. \quad (3.26)$$

They appear in eq. (3.21) because of the time dependence of the projector π . In general, such terms lead out of the operator space into which π projects.

In the strict adiabatic limit (small \dot{R}), one could neglect these terms because they are at least of second order in \dot{R} . However, we are interested in applying the theory also for the approach phase, where $|\nabla V|$ becomes large at the surface, so that $\dot{R}(|\nabla V|/V)\Delta$ may become large. At the same time $\langle v \rangle \Delta$ remains small due to the large energy denominator in the definition of v . Therefore, we shall not neglect the terms (3.26), but introduce a further approximation to calculate them. We shall show that because of the localized nature of ∇V , its matrix elements can be approximately factored, such that

$$\langle k\kappa | \nabla V(R') | l\lambda \rangle \approx \frac{g_{kl}(R')}{g_{kl}(R)} \langle k\kappa | \nabla V(R) | l\lambda \rangle. \quad (3.27)$$

We can justify this approximation in two extreme situations. As long as the two reacting nuclei do not closely overlap, only the tails of the wave functions overlap with the strongly peaked force ∇V . There, the wave functions behave roughly as $|l\lambda\rangle \sim \exp(-\alpha_l R)$, independent of the index within the (relatively narrow) bin. Under these conditions, $g_{kl}(R)$ would be roughly proportional to $\exp(-(\alpha_l + \alpha_k)R)$.

Similarly, if the nuclei penetrate deeply, the wave functions would resemble plane waves, and the relation (3.27) will hold with $g_{kl}(R) \sim \exp(i(k_l - k_k)R)$. (This situation is realized in the model of a piston moving in a Fermi gas²³), and expresses the fact that the force acting on the piston is due to the recoil of the gas particles from the piston²⁴). The approximation (3.27) implies, therefore, that we interpolate the factorization property between the two extremes mentioned above.

In the appendix we show that because of (3.27)

$$v_{k\kappa, l\lambda}(t') \approx \frac{G_{kl}(t')}{G_{kl}(t)} v_{k\kappa, l\lambda}(t), \quad (3.28a)$$

$$\frac{d}{dt} v_{k\kappa, l\lambda}(t) \approx \frac{\dot{G}_{kl}(t)}{G_{kl}(t)} v_{k\kappa, l\lambda}(t), \quad (3.28b)$$

with

$$G_{kl}(t) = \dot{R}g_{kl}(R), \quad G_{kl}^*(t) = G_{lk}(t). \quad (3.29)$$

This *ansatz* for v leads to

$$f_{kl}(t) = \left(\frac{\dot{G}_{kl}(t)}{G_{kl}(t)} \right) S_{kl}(t); \quad (3.30)$$

f_{kl} is given in terms of the coherences, and eqs. (3.20) and (3.21) become closed.

We now come back to the discussion of the correlation functions (3.22)–(3.25). One can easily check that

$$I_{kl}(t, t') = \frac{G_{kl}(t)}{G_{kl}(t')} \tilde{I}_{kl}(t, t'), \quad (3.31a)$$

$$\overset{(c)}{I}_{kl}(t, t') \equiv \frac{\partial}{\partial t} \tilde{I}_{kl}(t, t'), \quad (3.31b)$$

$$J_{kl}(t, t') = \int_{t'}^t dt'' \tilde{I}_{kl}(t, t'') = \int_{t'}^t dt'' \frac{G_{kl}(t'')}{G_{kl}(t)} I_{kl}(t, t''), \quad (3.31c)$$

$$\overset{(c)}{J}_{kl}(t, t') = \int_{t'}^t dt'' \frac{G_{kl}(t'')}{G_{kl}(t)} \overset{(c)}{I}_{kl}(t, t''). \quad (3.31d)$$

We define

$$\left(\frac{\overset{(c)}{I}_{kl}(t, t')}{I_{kl}(t, t')} \right)_\Delta \equiv \frac{1}{\Delta} \int_0^\Delta \frac{\overset{(c)}{I}_{kl}(t, t')}{I_{kl}(t, t')} dt' \equiv -i\omega_{kl}(t) - \Gamma_{kl}(t). \quad (3.32)$$

The postulated separability of time scales ($\tau^d \gg \Delta \gg \tau^c$) has two important consequences: (a) The functions $\omega_{kl}(t)$ and $\Gamma_{kl}(t)$ are almost constant over a time interval of the order Δ . (b) The definition (3.32) is independent of the exact value of Δ as long as $\tau^d \gg \Delta \gg \tau^c$. We can turn the argument around and state that if ω_{kl} and Γ_{kl} as functions of Δ (for a given t) do not show a “plateau” behaviour for $\tau^d \gg \Delta \gg \tau_c$, the assumption of separation of time scales is not fulfilled and the theory is not applicable²²). The functions $\omega_{kl}(t)$ and $\Gamma_{kl}(t)$ obey the symmetry relations

$$\omega_{kl} = -\omega_{lk}, \quad \Gamma_{kl} = \Gamma_{lk}. \quad (3.33)$$

We define

$$\eta_{kl} = -i\omega_{kl} - \Gamma_{kl} + (\dot{G}_{kl}/G_{kl})^*, \quad (3.34)$$

with

$$\eta_{kl}^* = \eta_{lk}. \quad (3.35)$$

Using these definitions and the relations (3.31), we get the final form for equations (3.20)–(3.21).

$$\dot{P}_l = 2 \operatorname{Im} \sum_k S_{kl}, \quad (3.36)$$

$$i\dot{S}_{kl} = i\eta_{kl}S_{kl} + \gamma_{kl}^2(P_l/d_l - P_k/d_k), \quad S_{lk} = S_{kl}^*. \quad (3.37)$$

We now consider the equations which govern the excitation of the special states $|I\rangle$. We treat these as “bins” containing only one state. Since we are not losing information about these states, they will obey exact von Neumann equations within the subspace, except for terms that connect them to the statistical bins. Thus we have

$$\dot{\rho}_{II} = -i \sum_J (v_{IJ}\rho_{JI} - \rho_{IJ}v_{JI}) + 2 \operatorname{Im} \sum_k S_{kI}, \quad (3.38)$$

$$i\dot{\rho}_{IJ} = \hbar^{-1}(E_I - E_J)\rho_{II} + \sum_L (v_{IL}\rho_{LJ} - \rho_{IL}v_{LJ}), \quad I \neq J. \quad (3.39)$$

In eqs. (3.36) and (3.37) we have used our convention that the indices k, l etc. refer to both statistical bins and state “bins”. To make the distinction explicit we rewrite these equations as follows:

$$\dot{P}_l = 2 \operatorname{Im} \left(\sum_k S_{kl} + \sum_I S_{II} \right), \quad (3.40)$$

$$i\dot{S}_{II} = -\gamma_{II}^2(\rho_{II} - P_l/d_l) + i\eta_{II}S_{II}, \quad (3.41)$$

$$i\dot{S}_{kl} = -\gamma_{kl}^2(P_k/d_k - P_l/d_l) + i\eta_{kl}S_{kl}. \quad (3.42)$$

Eqs. (3.38)–(3.42) are our final result for the reduced equations of the internal motion.

The variation of the collective coordinates determines how all the coefficients in the REM depend on time. In turn, the collective coordinates, through eq. (2.2), depend on an induced force which is given in terms of the coherences. This force is discussed in detail in sect. 4. The equation of motion for R , to be solved simultaneously with (3.38)–(3.42), is

$$M\ddot{R} = -\nabla U_{\text{opt}} + (2\hbar/\dot{R}) \sum_{n>m} (\omega_{nm} \operatorname{Im} S_{nm} - \Gamma_{nm} \operatorname{Re} S_{nm}) - \sum_{I,J} \rho_{IJ} (\partial V/\partial R)_{JI}. \quad (3.43)$$

It is to be understood that the sum over n and m includes mixed terms involving one index of the special states with one index of the statistical states.

The initial conditions for the entire set of coupled equations are, at $t \rightarrow -\infty$,

$$\begin{aligned} R &= \text{large}, \quad \dot{R} = (2E/M)^{1/2}, \\ \rho_{IJ} &= \delta_{I0}\delta_{J0}, \quad P_n = 0, \quad S_{nI} = S_{In} = S_{nm} = 0. \end{aligned} \quad (3.44)$$

The equations which describe the excitation of the special states are written directly in terms of the density matrix elements ρ_{IJ} and the probabilities $P_I = \rho_{II}$. Because of the initial conditions (3.44), eqs. (3.38)–(3.39) describe the initial stages of the collision.

If we disregard the last term (3.38), we obtain the usual quantum-mechanical description of a system driven by an external time dependent field. These equations are written in the adiabatic *representation*, but the adiabatic *approximation* is not imposed. The term $2 \operatorname{Im} \sum_k S_{kI}$ transfers flux from the $|I\rangle$ subspace to the statistical

space described by the bins. Comparing (3.38) and (3.40) we see that

$$\frac{d}{dt} \sum_I P_I = -\frac{d}{dt} \sum_I P_I. \quad (3.45)$$

The flow of probability flux is directed from the $|I\rangle$ space to the statistical space because of the large phase space available in the latter. Thus, after some time the $|I\rangle$ subspace will be emptied ($\sum_I P_I \rightarrow 0$) and the collision will be controlled by equations (3.40)–(3.42) which describe the statistical part of the process.

The set (3.40)–(3.42) drives the system to statistical equilibrium, which is reached when the populations P_n become proportional to the phase space factors d_n . The approach to equilibrium can be better investigated by solving eq. (3.42) and substituting it in (3.40). After a time, t_s , at which the “source” term $\sum_I S_{II}$ in (3.40) can be neglected [this is somewhere in the region of close contact, where we may also neglect $\dot{G}_{kl}(t)$ defined by eq. (3.29)], we obtain:

$$S_{mn} = \int_{t_s}^t \exp[\eta_{mn}(t-\tau)] i\gamma_{mn}^2(\tau) (P_m/d_m - P_n/d_n)_\tau d\tau. \quad (3.46)$$

Hence,

$$\dot{P}_n = 2 \sum_m \operatorname{Re} \int_{t_s}^t d\tau \exp[\eta_{mn}(t-\tau)] \gamma_{mn}^2(\tau) (P_m/d_m - P_n/d_n)_\tau. \quad (3.47)$$

Eq. (3.47) are the generalized rate equations discussed in sect. 2. In a pre-equilibrium situation, the differences $P_m/d_m - P_n/d_n$ vary on a slower time scale than τ^d , the decay constant of $\exp[\eta_{mn}(t-\tau)]$. One can introduce the Markovian approximation discussed in sect. 2 to obtain a proper rate equation and reach a complete analogy between the two descriptions.

We thus see that the present theory supplies us with a uniform and self-consistent description of the DIC process. It provides a natural scheme in which the coherent driving which characterizes the initial stages of the collision turns smoothly into a statistical description of a quasi-equilibrium situation. The time resolution which is dictated by the fast processes in the approach phase is kept throughout.

4. The induced force

In this section we discuss the induced force $\langle F \rangle$ (eq. (2.2)) in terms of the coherences S_{nm} . We demonstrate the procedure for a single degree of freedom. Looking back at the definition (2.8) of $v_{n\nu,m\mu}$, one can easily see that

$$\begin{aligned} \langle F \rangle &= -\operatorname{tr} \left[\rho \frac{1}{R} \frac{\partial V}{\partial t} \right] = \sum_{n\nu} \langle n\nu | F | n\nu \rangle \rho_{n\nu,n\nu} \\ &\quad - (i/R) \sum'_{n\nu,m\mu} v_{n\nu,m\mu}^* \rho_{n\nu,m\mu} (E_{n\nu} - E_{m\mu}) \\ &\quad + \sum_{I,J} \langle I | F | J \rangle \rho_{IJ} - (2i/R) \sum_{n\nu,I} \operatorname{Re} (v_{n\nu,I}^* \rho_{n\nu,I} (E_{n\nu} - E_I)). \end{aligned} \quad (4.1)$$

The first term is an “adiabatic” force. It is that part of the force that is not connected to transitions or excitations of the system and it should be identified with the negative gradient of the real optical potential $-\nabla U_{\text{opt}}$. One should not call it “conservative” since it is not necessarily a function of R alone. The remaining terms F_D , are connected with the excitation of the system and contain the dissipative force. We can express F_D in terms of the coherence operators.

$$\begin{aligned} F_D &= -(i\hbar/\dot{R}) \sum_{nm} \gamma_{nm} \langle A(n, m) | L^{(0)} | \rho \rangle \\ &= -(i\hbar/\dot{R}) \sum_{nm} \langle A(n, m) | (L - L^{(1)}) | \rho \rangle. \end{aligned} \quad (4.2)$$

Now,

$$-i \langle A(n, m) | L | \rho \rangle = \dot{S}_{nm} - (\dot{G}_{nm}/G_{nm})^* S_{nm}, \quad (4.3)$$

$$\langle A(n, m) | L^{(1)} | \rho \rangle = \langle A(n, m) | L^{(1)} U(\pi U \pi)^{-1} \pi | \rho \rangle, \quad (4.4)$$

which is, to lowest order in $L^{(1)}$,

$$\langle A(n, m) | L^{(1)} | \rho \rangle = \gamma_{nm}^2 (P_m/d_m - P_n/d_n). \quad (4.5)$$

The contribution of these terms vanish after summing over n and m because $\gamma_{nm}^2 = \gamma_{mn}^2$. Hence we have

$$\begin{aligned} F_D &= (\hbar/\dot{R}) \sum_{nm} (\dot{S}_{nm} - (\dot{G}_{nm}/G_{nm})^* S_{nm}) \\ &= -(\hbar/\dot{R}) \sum_{nm} (i\omega_{nm} + \Gamma_{nm}) S_{nm}. \end{aligned} \quad (4.6)$$

Using $\omega_{nm} = -\omega_{mn}$, $\Gamma_{nm} = \Gamma_{mn}$, $S_{nm} = S_{mn}^*$ the relation (4.1) reads finally:

$$\begin{aligned} \langle F \rangle &= -\nabla U_{\text{opt}} + (2\hbar/\dot{R}) \sum_{n>m} (\omega_{nm} \text{Im } S_{nm} - \Gamma_{nm} \text{Re } S_{nm}) \\ &\quad + \sum_{I \neq J} \langle I | F | J \rangle \rho_{JI} + (2\hbar/\dot{R}) \sum_{n,I} (\omega_{nI} \text{Im } S_{nI} - \Gamma_{nI} \text{Re } S_{nI}). \end{aligned} \quad (4.7)$$

The situation becomes somewhat more complicated when the number of collective variables exceeds one. The coherences are defined with respect to the operator $\dot{V} = R \cdot \nabla V$, and one cannot isolate the components of the force vector from the expectation values of the scalar \dot{V} . One overcomes this difficulty in the following way. Writing

$$\dot{V} = \sum_{i=1} \dot{R}_i \frac{\partial}{\partial R_i} \hat{V}, \quad (4.8)$$

we can define the operators

$$v_{nv,m\mu}^{(j)} = \begin{cases} i\dot{R}_j \frac{\langle nv | \partial \hat{V} / \partial R_j | m\mu \rangle}{E_{nv} - E_{m\mu}}, & nv \neq m\mu \\ 0, & nv = m\mu. \end{cases} \quad (4.9)$$

We then define the partial coherence operators $A^{(j)}(n, m)$ in terms of the $v^{(j)}$ and write the REM for the populations and the newly-defined coherences $S_{nm}^{(j)}$. The only new aspect in the derivation of the REM is the fact that the $A^{(j)}(n, m)$ for the same n, m but different j are not necessarily orthogonal tetrads. This turns out to be a minor technical difficulty¹⁸⁾. It has an important consequence for the physical picture, since it brings about the correlations between the various components of the induced force which, in the multi-dimensional case, relate to the velocity vector via the friction tensor.

We now demonstrate that the induced force in (4.7) reduces to that obtained in perturbation theory²⁴⁾ and linear-response theory⁶⁾. Nothing need be said about the force coming from the I, J space. It is treated exactly. For simplicity we discuss only the term

$$F_D(t) = (2\hbar/\dot{R}) \sum_{n>m} (\omega_{nm} \operatorname{Im} S_{nm} - \Gamma_{nm} \operatorname{Re} S_{nm}). \quad (4.10)$$

The formal solution of the REM for the coherences S_{nm} (3.41) is

$$S_{nm}(t) = i \int_{t_0}^t \exp \left(\int_{t'}^t \eta(\tau) d\tau \right) \gamma_{nm}^2(t') (P_n/d_n - P_m/d_m)_{t'} dt'. \quad (4.11)$$

$\eta(t)$ has been defined by

$$\eta_{nm}(t) = -i\omega_{nm} - \Gamma_{nm} + (\dot{G}_{nm}/G_{nm})^*. \quad (4.12)$$

We can write

$$\int_{t'}^t \eta_{nm}(\tau) d\tau = (-i\omega_{nm} - \Gamma_{nm})(t - t') + \ln (G_{nm}^*(t)/G_{nm}^*(t')), \quad (4.13)$$

i.e.

$$S_{nm}(t) = i \int_{t_0}^t \exp (-i\omega_{nm} - \Gamma_{nm})(t - t') \sum_{\nu\mu} v_{n\nu,m\mu}^*(t) v_{n\nu,m\mu}(t') (P_n/d_n - P_m/d_m)_{t'} dt'. \quad (4.14)$$

In other words, the function $\tilde{I}_{nm}(t, t')$ appears here. If we assume that \tilde{I}_{nm} can be replaced by $\gamma_{nm}^2 \exp (-i\omega_{nm} - \Gamma_{nm})(t - t')$, which is an approximation consistent with the philosophy of this paper, we find

$$F_D = (1/\dot{R}(t)) \int_{t_0}^t \sum_{n\nu,m\mu} (E_{n\nu} - E_{m\mu}) v_{n\nu,m\mu}^*(t) \exp \left(-\frac{i}{\hbar} \int_{t'}^t (E_{n\nu}(t'') - E_{m\mu}(t'')) dt'' \right) \\ \times v_{n\nu,m\mu}(t') (P_n/d_n - P_m/d_m)_{t'} dt' + \text{c.c.} \quad (4.15)$$

Using the definition of v^* [eq. (2.8)], and of ω_{nm} and Γ_{nm} [eq. (3.30)], we get

$$F_D = \frac{1}{\dot{R}(t)} \int_{t_0}^t \sum_{n\nu,m\mu} \langle n\nu | \dot{V}(t) | m\mu \rangle \frac{\exp(-(i/\hbar) \int_{t'}^t (E_{n\nu}(t'') - E_{m\mu}(t'')) dt'')}{E_{n\nu} - E_{m\mu}} \langle m\mu | \dot{V}(t') | n\nu \rangle \\ \times (P_n/d_n - P_m/d_m)_{t'} dt' + \text{c.c.} \quad (4.16)$$

Once the system has developed into the quasi-equilibrium phase, the factor $(P_n/d_n - P_m/d_m)_t$ becomes almost constant on the time scale of the correlation function with which it is convoluted in (4.16). It can be taken out of the integral and evaluated at $t' = t$. We thus obtain the important result that when the internal system reaches equilibrium, with $(P_n/d_n = P_m/d_m)$, the friction force vanishes, as it must since there is no net transfer of energy among the bins.

Once one assumes the applicability of the Markovian approximation discussed above, the induced force (4.16) coincides with the expression one would obtain from perturbation theory²⁴⁾, in which one assumes that the internal density matrix is given as $\rho_{n\nu,m\mu} = \delta_{n\nu,m\mu} P_n/d_n$.

5. Summary

The basic ingredient of a statistical description of deeply inelastic collisions is the existence of a hierarchy of time scales in the process. From trajectory models of DIC we know that^{1,25)}:

(a) There is a fast excitation process in the ingoing channel. Within the time $t_{\text{rad}} = 10^{-22}$ s a considerable amount of energy (up to about 100 MeV) is transferred from the relative radial motion into intrinsic excitations. The relative motion suffers a strong frictional slowing down. Within the same time the $(Z - N)$ degree of freedom equilibrizes by means of the exchange of a few nucleons.

(b) After close contact is established, the system continues to transfer energy into the intrinsic system and to exchange mass. This phase of the reaction is usually called the contact phase, and its typical duration t_{cont} , is a few 10^{-21} s. Within t_{cont} the two nuclei become strongly deformed until they separate again.

The contact phase is commonly described by transport theories^{2,4,5,6)} which consider the internal system to be close to equilibrium. That is, the internal density matrix $\hat{\rho}$ is assumed to deviate only slightly from a microcanonical^{2,5)} or a canonical^{4,6)} distribution. (In our language, the equilibrium $\hat{\rho}$ referred to above, is given by the populations alone – diagonal matrix elements of $\hat{\rho}$ in the energy-representation.) In the initial stage of the collision this is certainly not true. The time t_{eq} it takes the system to reach its internal equilibrium is of the same order as t_{rad} . During the time t_{rad} coherent excitations of the internal system are important, and the intrinsic density is not described by the populations alone, since important information is contained in the off-diagonal elements of $\hat{\rho}$.

In the present paper we have shown how the statistical and quantum mechanical aspects of the processes which occur during the approach and the contact phases can be uniformly described by a theory that treats the populations and coherences explicitly on the same footing. The populations were defined by the mean value of the diagonal matrix elements of $\hat{\rho}$ within a given energy bin, and the coherences are the averages of the off-diagonal elements of $\hat{\rho}$ between two bins, weighted by the corresponding transition matrix elements of the coupling v .

In deriving the reduced equations of motion (REM) (3.16), we used a new method¹⁸), and the resulting equations do not contain convolution in time. This is formally different from the more conventional treatments which yield integro-differential REM¹⁵). A detailed discussion and comparison of both approaches can be found elsewhere²⁰). The following points should be made here. (a) Both reduction schemes are exact and are equivalent to the complete von Neumann equation, provided that the kernels are evaluated to all orders. Once approximations are made, they may, however, yield very different results, and then one scheme may become more advantageous. The actual choice depends to a large extent on the statistical properties of our system. It was shown²⁸) that the present REM (without time convolution) are particularly useful for ensembles with gaussian properties, since then, higher order terms in the results vanish identically. (b) Technically, the evaluation of the results in the present formalism involves the perturbative expansion of $U(t, t_0) = \exp(-iL(t - t_0))$, whereas in the other scheme one needs $\exp(-i(1 - \pi)L(1 - \pi)(t - t_0))$. For our choice of dynamical variables, which include coherences, the former expansion is straightforward, whereas the latter is very complicated. This is therefore the natural reduction scheme for our purpose.

The main assumption of the present work is that a *separation of time scales* exists with respect to the internal motion of the system. We have introduced two characteristic times: the dephasing time τ^d which measures the rate of decay of the coherences, and τ^c which measures the lifetime of excitations into the irrelevant space of dynamical operators projected by $(1 - \pi)$. We require the following hierarchy to hold:

$$t_{\text{cont}}(>\tau^P) > t_{\text{rad}}(\equiv t_{\text{eq}} = \tau^d) \gg \tau^c, \quad (5.1)$$

where τ^P is the rate of change of the populations during the contact phase. It is related to τ^d through $\tau^P = (\Omega^2 \tau^d)^{-1}$ [eq. (2.16)]. We chose to introduce the Markovian approximation on a time scale Δ , intermediate between τ^d and τ^c . If we solve (3.42) for the coherences and substitute the result into the population equations (3.40), then these are non-Markovian. They become Markovian only over a time scale larger than τ^d . The equations are identical to conventional transport theories valid for the contact phase. Because τ^d is the dephasing time of the internal motion it is of the order of $\hbar/(a \text{ few MeV})$ (the typical p-h excitation energy), i.e. $\tau^d \sim 10^{-22} \text{ s}$, and thus similar to τ_{rad} . By including the explicit dynamics of the coherences we are able to describe relaxation processes on a finer time scale $\Delta < \tau^d$. Thus, we hope our theory is able to describe the fast processes within the initial stage discussed under (a) above.

Our REM turn out to be the exact von Neumann equation for $\hat{\rho}$ within the Hilbert space of special states $|I\rangle$. Therefore, we have an exact description of the very early coherent excitations. As the reaction goes on, flux will be shifted from

the $|I\rangle$ states into the higher lying excited bins of states. The populations P_n of the higher bins start to grow via the coherences S_{In} , the ν -averaged transition matrix from the $|I\rangle$ to the $|n\nu\rangle$ states. Finally, the populations of the bins equilibrate towards equipartition within the phase space, $P_n/d_n = P_m/d_m$, due to the coarse-grained coherent transitions S_{nm} between the statistical bins. This is the stochastic regime, valid in the contact phase. We show that our REM switch smoothly to the rate equations considered in conventional transport theories^{2,4,5}). The transition from the coherent reversible description of the early process to the irreversible stochastic description of the latter phases is obtained through an increasing loss of information. The latter is due to the coarse graining of the higher part of the spectrum, i.e. it is due to our considering reduced equations of motion for $\pi\hat{\rho}$ only, and also due to coarse graining in time by averaging the relevant correlation functions over time Δ .

Our REM (3.38)–(3.43) conserve probability, but in contrast with the basic equations (2.1)–(2.2), do not guarantee the conservation of energy and angular momentum. Violations of conservation laws may indicate that the populations and the coherences do not exhaust the entire space of slow variables. This point will be discussed in a forthcoming publication where a new energy conserving REM will be derived.

The basic microscopic input consists of the mean matrix elements γ_{nm}^2 and the ratios $I_{nm}(t)/\langle I_{nm}(t)\rangle$. Both may be calculated in a first application by using average matrix elements, for example, from the work of ref.²⁷). Restricting ourselves at the beginning to inelastic excitations only (neglecting mass transfer) the important question may be answered as to whether the rapid transfer of approximately 100 MeV is possible during the initial (coherent) stage of the collision. In such calculations, the onset of the statistical phase would be characterized by: (a) the $\dot{S}_{nm}(t)$ approaching zero, and (b) the P_n approaching a canonical distribution with a “temperature” determined by the energy lost from the mean collective trajectory.

In this paper the collective and the internal coordinates are both treated dynamically in a self-consistent way. However, we have neglected correlations between the intrinsic density distributions $\hat{\rho}(t)$ and the distribution $\sigma(RP_t)$ of the collective degrees of freedom, i.e. we have taken the total density $\hat{D}(R, P, t)$ of the system to be separable at any time, $\hat{D}(R, P, t) = \hat{\rho}(t)\sigma(R, P, t)$. In paper II we will relax this limitation by including the correlations $\hat{D}(R, P, t) - \hat{\rho}(t)\sigma(R, P, t) = \hat{\Delta}(R, P, t)$ via a generalized Fokker–Planck equation.

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Appendix

Here we consider the derivation of eq. (3.28) for $dv_{n\nu,m\mu}/dt$. We have

$$\begin{aligned}
 \frac{d}{dt} v_{n\nu,m\mu}(t) &= \frac{d}{dt} \frac{i\langle n\nu | \dot{V}(t) | m\mu \rangle}{E_{n\nu}(t) - E_{m\mu}(t)} \\
 &= \frac{i}{E_{n\nu} - E_{m\mu}} \left\{ \langle n\nu | \ddot{V} | m\mu \rangle + \sum_{l\lambda \neq n\nu, m\mu} \langle n\nu | \dot{V} | l\lambda \rangle \langle l\lambda | \dot{V} | m\mu \rangle \right. \\
 &\quad \times \left[\frac{1}{E_{m\mu} - E_{l\lambda}} + \frac{1}{E_{n\nu} - E_{l\lambda}} \right] \\
 &\quad \left. + 2 \frac{\langle n\nu | \dot{V} | m\mu \rangle}{E_{n\nu} - E_{m\mu}} (\langle m\mu | \dot{V} | m\mu \rangle - \langle n\nu | \dot{V} | n\nu \rangle) \right\}. \quad (\text{A.1})
 \end{aligned}$$

The last two terms come from the differentiation of the energy-denominator in $v_{n\nu,m\mu}$.

Because of our assumption of random phases of the v 's [eq. (3.18)] the second term in (A.1) vanishes. Also the third term can be neglected, because it is of second order in $\dot{V}/\Delta E$; its contribution to the REM for S_{nm} is of the form $(\dot{V}/\Delta E)S$ and thus of one order higher than the other terms in eq. (3.41).

There remains

$$\frac{d}{dt} v_{n\nu,m\mu}(t) = \frac{i\langle n\nu | \ddot{V} | m\mu \rangle}{E_{n\nu} - E_{m\mu}} = \frac{d(\dot{R}g_{nm}(R))/dt}{\dot{R}g_{nm}(R)} \frac{i\langle n\nu | \dot{V} | m\mu \rangle}{E_{n\nu} - E_{m\mu}} = \frac{\dot{G}_{nm}(t)}{G_{nm}(t)} v_{n\nu,m\mu}(t), \quad (\text{A.2})$$

the result quoted in (3.28). In the last step we have pulled one time differentiation out of the matrix elements, using the fact that differentiating the state $|m\mu\rangle$ introduces random contributions like the second term in (A.1).

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