Final-State Effects on Thermal-Neutron Scattering at High-Energy Transfer*

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An approximation is developed for nuclear scattering of neutrons from a target of spinless particles of a single isotope. It has the correct limit for large momentum transfer, the impulse approximation, and is calculable in practice if the single-particle momentum distribution, the off-diagonal two-particle density matrix, and the two-body interaction potential of the target atoms are known. An intended application of the theory is to neutron scattering from liquid helium in the moderately-high momentum-transfer region ($\vec{k} \approx 15$ Å^{-1}) presently accessible with reactor neutrons.

I. INTRODUCTION

The inelastic scattering cross section for nuclear scattering of neutrons from a target composed of spinless particles of a single isotope is given in the Born approximation[1] by

$$\frac{d^2\sigma}{d\Omega\, d\epsilon_f} = \frac{\sigma_b k_f}{4\pi\hbar k_i} S(\vec{k}, \omega), \tag{1}$$

where $\hbar\vec{k} = \hbar\vec{k}_i - \hbar\vec{k}_f$ is the momentum transferred to the target, $\hbar\omega = \epsilon_i - \epsilon_f$ is the energy transfer, and σ_b is the neutron-target-particle total cross section. The dynamic structure factor $S(\vec{k}, \omega)$ is the Fourier transform of the density-density correlation function $S(\vec{k}, t)$ for a system with N particles,

$$2\pi S(\vec{k}, \omega) = \int_{-\infty}^{\infty} dt\, e^{-i\omega t} S(\vec{k}, t), \tag{2}$$

where

$$NS(\vec{k}, t) = \sum_{j,l} \langle e^{-i\vec{k}\cdot\vec{r}_l(0)} e^{i\vec{k}\cdot\vec{r}_j(t)} \rangle . \tag{3}$$

Equation (3) contains the Heisenberg operator $\vec{r}_j(t)$ defined for all j and t by

$$\vec{r}_j(t) = e^{iHt/\hbar} \vec{r}_j e^{-iHt/\hbar} . \tag{4}$$

For asymptotically large values of momentum transfer, the dynamic structure factor may be realistically evaluated in the impulse approximation. This approximation is obtained from Eq. (3) by replacing the Hamiltonian of the target by a free-particle Hamiltonian in Eq. (4),

$$H \to H_0 = \sum_i \frac{P_i^2}{2m}, \tag{5}$$

and by discarding coherent terms, $j \neq l$, in the double sum in Eq. (3). To evaluate this approximation for a particular target, the only information required is the single-particle momentum distribution n_p of the target.

A main deficiency of the impulse approximation is that in general there is no rigorous method to determine the values of k for which it is applicable. A second potential deficiency is that for some physically interesting systems it may be impractical to perform experiments at the large momentum transfers required for the use of the impulse approximation. As a possible remedy for these problems, an approximation is developed here which should be valid at more moderate values of the momentum transfer. This approximation has the correct limit for large momentum transfer, the impulse approximation, and is calculable in practice if the single-particle momentum distribution, the off-diagonal two-particle density matrix, and the two-body interaction potential of the target particles are known. This paper deals with the details of this approximation. A subsequent paper[2] applies these results to a detailed analysis of an experimental investigation of the scattering of high-energy ($\epsilon_i \approx 200$ meV) neutrons from liquid helium, performed by Mook, Scherm, and Wilkinson.[3]

The form of the result may be motivated by writing the impulse approximation as

$$S_{\mathrm{IA}}(\vec{k}, \omega) = \sum_{\vec{p}} n_{\vec{p}}\, \delta\left(\omega - \frac{\hbar k^2}{2m} - \frac{\vec{k}\cdot\vec{p}}{m}\right), \tag{6}$$

and ascribing the following picture to it. The neutron strikes a single particle in the target. The struck particle has a momentum $\vec{p}$ initially and the collision is elastic and conserves the total momentum of the neutron and target particle. The δ function in Eq. (6) is the mathematical statement that kinetic energy and momentum are conserved in this two-body collision. This is true only to the extent that the target-particle interactions are negligible. At a more modest momentum transfer the target interactions will have some effect. Conceptually, at this lower value of $\vec{k}$, one could still picture the neutron striking a single particle and replace the remaining particles of the target by an effective potential. The presence of this potential would remove the requirement that the neutron-particle collision

conserve momentum and would allow for inelastic processes. A momentum state $\vec{p}$ would then contribute to the scattering not only at the precise value of the energy transfer ω which satisfies $\omega - \hbar k^2/2m - \vec{k}\cdot\vec{p}/m = 0$, but for all ω for which $\omega - \hbar k^2/2m - \vec{k}\cdot\vec{p}/m$ is small, i.e., almost-elastic collisions which approximately satisfy conservation of the neutron-particle momentum. In mathematical terms the δ function in Eq. (6) would be replaced by a finite-width function $R(\vec{k}, \omega - \hbar k^2/2m - \vec{k}\cdot\vec{p}/m)$ which describes the effect of final-state interactions. The incoherent contribution to the dynamic structure factor would become

$$S_i(\vec{k}, \omega) = \sum_{\vec{p}} n_{\vec{p}} R\left(\vec{k}, \omega - \frac{\hbar k^2}{2m} - \frac{\vec{k}\cdot\vec{p}}{m}\right), \tag{7}$$

where the width of the function R would depend on the momentum transfer k and on the properties of the target; for example, the interaction potential of the target particles. The width of R should decrease as k increases, approaching a δ function. Its width should also decrease if the interactions between the target particles were to weaken.

In Sec. II an expression is developed for the function R which is formally exact for all values of momentum and energy transfer. A complete evaluation of R is in general no more tractable than an evaluation of a more conventional expression for $S(\vec{k}, \omega)$, e.g., Eq. (3), but the exact form of R suggests approximations which are potentially manageable for many-body systems at moderate transfers. Roughly the applicability of the final approximation for R could be checked by comparing the width of the function R to that of the single-particle momentum distribution n_p. If the width of R were appreciably greater than n_p use of the approximation would be questionable.

II. DERIVATION OF THE EFFECT OF FINAL-STATE INTERACTIONS

Although the discussion in the Introduction centered on the incoherent-scattering contribution to the dynamic structure factor [$j = l$ terms in Eq. (3)], the derivation of the function R will be performed with the coherent contributions, corresponding to the $j \neq l$ terms in Eq. (3), included. The desired form of the structure factor, Eq. (7), can then be obtained from the final formulas by discarding the coherent terms, a procedure justified when considering scattering from a liquid at large momentum transfer.

Since the intended application of this work is to neutron scattering at large momentum and energy transfers, an attempt will be made to motivate the formal manipulations by picturing the scattering as occurring between a neutron and the jth target particle. The procedure begins by noting that the Hamiltonians in Eq. (4) which govern the time dependence of $S(\vec{k}, t)$ contain no explicit recognition that a scattering event has changed the momentum of the jth particle. To incorporate the modified momentum and kinetic energy of the jth particle, the right-hand side of Eq. (4) is multiplied by unity in the form $e^{i\vec{k}\cdot\vec{r}_j} e^{-i\vec{k}\cdot\vec{r}_j}$ from the left and then inserted into Eq. (3) (putting $\hbar = 1$):

$$NS(\vec{k}, t) = \sum_{j,l} \langle e^{i\vec{k}\cdot(\vec{r}_j - \vec{r}_l)} e^{-i\vec{k}\cdot\vec{r}_j} e^{iHt} e^{i\vec{k}\cdot\vec{r}_j} e^{-iHt} \rangle . \tag{8}$$

We now make use of the identity

$$e^{-i\vec{k}\cdot\vec{r}_j} e^{iHt} e^{i\vec{k}\cdot\vec{r}_j} = e^{iH't},$$

where

$$H' \equiv H(\vec{r}_1, \ldots, \vec{r}_N; \vec{p}_1, \ldots, \vec{p}_j + \vec{k}, \ldots, \vec{p}_N),$$
$$H' = H + \omega_k + L_j, \tag{9}$$

with $\omega_k = k^2/2m$, $L_j = \vec{k}\cdot\vec{p}_j/m$, under the assumption that the Hamiltonian contains only kinetic energy and a velocity-independent potential. The density-density correlation function now has the form

$$NS(\vec{k}, t) = e^{i\omega_k t} \sum_{j,l} \langle e^{i\vec{k}\cdot(\vec{r}_j - \vec{r}_l)} e^{i(H+L_j)t} e^{-iHt} \rangle . \tag{10}$$

In the modified Hamiltonian, Eq. (9), the momentum lost by the neutron is explicitly transferred to the jth particle in the target. The jth particle, carrying its modified momentum $\vec{p}_j + \vec{k}$, will move in the medium of its neighbors and encounter varying potential energies which will distort its trajectory from that of a free particle. The varying potential energy in its environment can be exhibited by using the relation

$$e^{iHt + iL_j t} = e^{iL_j t} T \exp\left[i \int_0^t H(\vec{r}_j - \vec{v}_k t')\, dt'\right], \tag{11}$$

where

$$\vec{v}_k = \vec{k}/m,$$

$$H(\vec{r}_j - \vec{v}_k t') \equiv H(\vec{r}_1, \ldots, \vec{r}_j - \vec{v}_k t', \cdots, \vec{r}_N; \vec{p}_1, \ldots, \vec{p}_N), \tag{12}$$

$$H(\vec{r}_j - \vec{v}_k t') = e^{-i\vec{p}_j\cdot\vec{v}_k t'} H e^{i\vec{p}_j\cdot\vec{v}_k t'},$$

and T is the time-ordering symbol. The Hamiltonian $H(\vec{r}_j - \vec{v}_k t')$, representing the motion of the struck particle, can be rewritten in terms of the original target Hamiltonian H as

$$H(\vec{r}_j - \vec{v}_k t') = H + U_j(v_k t'), \tag{13}$$

where

$$U_j(v_k t') = \sum_{\substack{m \\ (m \neq j)}} [V(\vec{r}_j - \vec{v}_k t', \vec{r}_m) - V(\vec{r}_j, \vec{r}_m)]. \tag{14}$$

The operator $U_j(v_k t)$ represents the change in the potential energy between the struck jth particle, as it travels along a straight-line trajectory with velocity $\vec{v}_k$, and the other target particles represented by m.

The density-density correlation function $S(\vec{k}, t)$ is now in the form

$$NS(\vec{k}, t) = e^{i\omega_k t} \sum_{j,l} \langle e^{i\vec{k}\cdot(\vec{r}_j - \vec{r}_l)} e^{i\vec{p}_j \cdot \vec{v}_k t} \times T \exp[iHt + i\int_0^t U_j(v_k t')\, dt'] e^{-iHt} \rangle . \tag{15}$$

The development to this point parallels our earlier treatment.[4] In the previous study we proceeded by expanding the T product. The first term in this expansion is the impulse approximation; the subsequent incoherent terms were shown to represent corrections to the impulse approximation ordered in increasing powers of $1/k$.

As noted in a previous paragraph, the natural picture to associate with the above procedure is of the struck particle traveling in a straight line. This suggests that difficulties may be encountered if the interaction between the target particles is strong for some configurations of the particles. For example, if the interaction contains a strong repulsive core, the second- and higher-order terms in the expansion of Eq. (15) have contributions from configurations in which the struck particle can pass arbitrarily close to another particle in the target without allowing either particle to readjust its position to avoid a close encounter. This can be avoided by noting that the unitary operators $e^{\pm iHt}$ in Eq. (15) temper the contributions from these unphysical encounters between the struck particle and its neighbors by allowing the particle coordinates to evolve in time. The time evolvement of the target-position coordinates can be made more explicit by use of the identity

$$T \exp[iHt + i\int_0^t U_j(v_k t')\, dt'] = T \exp[i\int_0^t e^{iH(t-t')} U_j(v_k t') e^{-iH(t-t')}\, dt'] e^{iHt} , \tag{16}$$

which can be interpreted as a resummation of the T product in Eq. (15). Since this identity was not obvious to us,[5] we give a proof of it in Appendix A. The exponent on the right-hand side of Eq. (16) is the integral of the difference in the potential the struck particle would have encountered as a typical target particle and the potential it does encounter as the atypical struck particle

$$e^{iH(t-t')} U_j(v_k t') e^{-iH(t-t')} = \sum_m{}' \tilde{U}_{j,m}(v_k t') = \sum_m{}' \{V[\vec{r}_j(t-t') - \vec{v}_k t', \vec{r}_m(t-t')] - V[\vec{r}_j(t-t'), \vec{r}_m(t-t')]\} , \tag{17}$$

where the term $m = j$ is deleted from the sum $\sum'$.

The function $S(\vec{k}, t)$ now has the form

$$NS(\vec{k}, t) = e^{i\omega_k t} \sum_{j,l} \left\langle e^{i\vec{k}\cdot(\vec{r}_j - \vec{r}_l)} e^{i\vec{v}_k t \cdot \vec{p}_j} \times T \exp\left(i\int_0^t dt' \sum_m{}' \tilde{U}_{j,m}(v_k t')\, dt' \right) \right\rangle . \tag{18}$$

For a realistic many-body system the detailed accounting for the time evolvement of all target coordinates contained in $\tilde{U}_{j,m}(v_k t')$ is an impossible task. In fact it is apparent that the defining equation for $S(\vec{k}, t)$, Eq. (3), appears very much simpler than the result expressed by Eq. (18). The apparent simplicity of Eq. (3) is deceptive. This becomes clear when one inserts for $\vec{r}_j(t)$ in Eq. (3) an exact result which follows from the Heisenberg equations of motion:

$$i\frac{\partial \vec{r}_j(t)}{\partial t} = [\vec{r}_j(t), H] = \frac{i\vec{p}_j(t)}{m};$$

$$i\frac{\partial \vec{p}_j(t)}{\partial t} = [\vec{p}_j, H] = -i\vec{\nabla}_j \sum_l{}' V(\vec{r}_j, \vec{r}_l) .$$

These yield the equation for the time dependence of the jth target coordinate,

$$\vec{r}_j(t) = \vec{r}_j(0) + \frac{\vec{p}_j(0)t}{m} - \frac{1}{m}\int_0^t dt'(t-t') \sum_l{}' \vec{\nabla}_j V[\vec{r}_j(t'), \vec{r}_l(t')] . \tag{19}$$

Inserting this expression for $\vec{r}_j(t)$ in Eq. (3), and accounting for the noncommutivity of the operators in Eq. (19), must yield an expression equivalent to Eq. (18). Incidentally, it is easy to see, by expanding Eqs. (3) and (18) in powers of k and comparing the terms linear in k, that (18) implies that $\vec{r}_j(t)$ is properly given by its exact value expressed in Eq. (19).

In Eq. (18) the impulse approximation still appears as an additive contribution to the dynamic structure factor. In order to obtain $S(\vec{k}, \omega)$ in the form given in Eq. (7) and to identify the function R which describes the effect of final-state interactions, a cumulantlike expansion[6] is performed. The appearance of the T product complicates the standard cumulant expansion procedure somewhat, so the details of this procedure are sketched in Appendix B. Applying this expansion yields

$$NS(\vec{k},t)=\tilde{S}_{IA}(\vec{k},t)\tilde{R}(\vec{k},t)\,, \tag{20}$$

where

$$N\tilde{S}_{IA}(\vec{k},t)=\sum_{j,l}\langle e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}e^{i\vec{v}_k t\cdot\vec{p}_j}\rangle e^{i\omega_k t} \tag{21}$$

and

$$\tilde{R}(\vec{k},t)=\exp[\omega_1+\omega_2+\cdots]\,, \tag{22}$$

where

$$\omega_1=-e^{i\omega_k t}\sum_{j,l,m}\frac{\langle e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}e^{i\vec{v}_k t\cdot\vec{p}_j}[1-T\exp[i\int_0^t\tilde{U}_{j,m}(v_k t')\,dt']]\rangle}{S_{IA}(\vec{k},t)}\,. \tag{23}$$

The form of the second term ω_2 in the exponent of Eq. (22) is given in Appendix B.

For systems where it is appropriate to discard the coherent-scattering contributions $j\neq l$, $\tilde{S}_{IA}(\vec{k},t)$ becomes $S_{IA}(\vec{k},t)$, the impulse approximation to the density-density correlation function

$$NS_{IA}(\vec{k},t)=\sum_j\langle e^{i\vec{v}_k t\cdot\vec{p}_j}e^{i\omega_k t}\rangle\,, \tag{24}$$

and $\tilde{R}(\vec{k},t)$ becomes

$$R(\vec{k},t)=\exp\left(\frac{-\sum_{j,m}\langle e^{i\vec{v}_k t\cdot\vec{p}_j}\{1-T\exp[i\int_0^t dt'U_{j,m}(\vec{v}_k t')]\}\rangle}{\sum_{j'}\langle e^{i\vec{v}_k t\cdot\vec{p}_{j'}}\rangle}+\cdots\right). \tag{25}$$

Equations (24) and (25) accomplish, at least formally, the objective stated in the Introduction, for when Eq. (20) (with $j\neq l$ terms discarded) is Fourier transformed to yield the incoherent contribution to the dynamic structure factor the result is

$$S_i(\vec{k},\omega)=\sum_{\vec{p}}n_{\vec{p}}R(\vec{k},\omega-\omega_k-\vec{p}\cdot\vec{v}_k)\,, \tag{26}$$

where the function $R(\vec{k},\omega-\omega_k-\vec{p}\cdot\vec{v}_k)$ is the Fourier transform of $R(\vec{k},t)$,

$$2\pi R(\vec{k},\omega-\omega_k-\vec{p}\cdot\vec{v}_k)=\int_{-\infty}^{\infty}dt\,e^{-i(\omega-\omega_k-\vec{p}\cdot\vec{v}_k)t}R(\vec{k},t)\,. \tag{27}$$

Equation (26) is still an exact result for the incoherent part of $S(\vec{k},\omega)$ and it is still intractable when applied to a realistic many-body system. For such systems it is necessary to apply some approximation. Our interest is in scattering at large neutron momentum transfers, and we seek an improvement over the impulse approximation, which sets the exponent $(\omega_1+\omega_2+\cdots)$ in Eq. (22) equal to zero for all times. The first term ω_1 in the exponent corresponds to the picture where the jth particle is struck by the neutron and then the jth particle scatters off the other particles in the target, each treated singly. The succeeding terms $\omega_2,\omega_3,\ldots$ in the exponent of Eq. (22) correspond to the scattering of the jth particle by clusters of two, three, ..., particles. One may expect that, at large momentum transfers, a meaningful correction to the impulse approximation results from neglecting all higher-order cumulants and retaining only the term ω_1 in the exponent of the function $\tilde{R}(\vec{k},t)$ in Eq. (22). This approximation neglects scattering of the jth particle by clusters of two or more target particles.

The calculation of $R(\vec{k},t)$ is now reduced to an evaluation of the cumulant ω_1. This appears to be a calculation of a two-body operator until one recognizes that the appearance of $\vec{r}_j(t-t')$ and $\vec{r}_m(t-t')$ in Eq. (25) leaves one with a problem of the same order of complexity as an exact calculation of $S(\vec{k},t)$. Further progress is made by noting that for large momentum transfers the time evolvement of $\vec{r}_j(t-t')-\vec{v}_k t'$ is dominated by $\vec{v}_k t'$ and therefore $e^{\pm iH(t-t')}$ in Eq. (17) may be treated cavalierly. One might, for example, replace the time evolvement generated by the full Hamiltonian with a time evolvement generated by an appropriate two-body Hamiltonian describing the struck jth particle and the mth particle with which it is interacting. An even simpler, though more drastic, approximation is obtained by completely neglecting the effect of the factors $e^{\pm iH(t-t')}$ in giving the target-particle locations a time dependence. This corresponds to the struck particle traveling along a straight line with the remaining particles frozen in their $t=0$ configuration, a process reminiscent of the eikonal approximation. Neglecting the time evolvement generated by H reduces the calculation to one in which the only information required

about the target is the one-particle off-diagonal density matrix, the two-particle off-diagonal density matrix, and the two-body potential. This follows from Eq. (25), which becomes

$$R(\vec{k},t)\cong\exp\frac{-\sum_{j,m}\langle e^{i\vec{v}_k\cdot\vec{p}_j t}(1-\exp\{i\int_0^t dt'[V(\vec{r}_j-\vec{v}_k t',\vec{r}_m)-V(\vec{r}_j,\vec{r}_m)]\})\rangle}{\sum_j\langle e^{i\vec{v}_k\cdot\vec{p}_j t}\rangle}. \tag{28}$$

The expectation value $\langle e^{i\vec{v}_k\cdot\vec{p}_j t}\rangle$ involves only a one-body operator and is thus reducible to a one-particle density matrix. This reduction is performed by averaging over a single state $\Psi_0(\vec{r}^N)$ for the target system; generalization to a canonical average is obvious. We have

$$\sum_j\langle e^{i\vec{v}_k\cdot\vec{p}_j t}\rangle=N\langle e^{i\vec{v}_k\cdot\vec{p}_j t}\rangle =N\int\Psi_0^*(r^N)e^{i\vec{v}_k\cdot\vec{p}_j t}\Psi_0(r^N)\,d\tau^N . \tag{29}$$

The operator $e^{i\vec{v}_k\cdot\vec{p}_j t}$ shifts the coordinate $\vec{r}_j$ appearing in $\Psi_0(r^N)$ by the amount $\vec{v}_k t$, and the integration over the coordinates $\vec{r}_2,\cdots,\vec{r}_N$ introduces the one-particle density matrix $\rho_1(\vec{r}_1,\vec{r}_1')$ defined by

$$\rho_1(\vec{r}_1,\vec{r}_1')=N\int\Psi_0^*(\vec{r}_1,\vec{r}_2,\cdots,\vec{r}_N)\times\Psi(\vec{r}_1',\vec{r}_2,\cdots,\vec{r}_N)d\tau_2\cdots d\tau_N . \tag{30}$$

The result is

$$\sum_j\langle e^{i\vec{v}_k\cdot\vec{p}_j t}\rangle=\int d\tau_1\,\rho_1(\vec{r}_1,\vec{r}_1+\vec{v}_k t)=\Omega'\rho_1(0,\vec{v}_k t), \tag{31}$$

where Ω' is the volume of the target system, and we have used translational invariance of the wave function Ψ_0 to obtain the last equality.

In a similar way, the expectation value in the numerator of the exponential in Eq. (28) involves only a sum of two-body operators and can be written in terms of a two-particle density matrix. The reduction is accomplished by writing the sum over j and m as $N(N-1)$ times the expectation value for a chosen pair, say, particles 1 and 2. This term is then

$$N(N-1)\int\Psi_0^*(r^N)e^{i\vec{v}_k\cdot\vec{p}_1 t}(1-\exp\{i\int_0^t[V(\vec{r}_1-\vec{v}_k t',\vec{r}_2)-V(\vec{r}_1,\vec{r}_2)]\,dt'\})\Psi_0(r^N)\,d\tau^N \tag{32}$$

$$=N(N-1)\int\Psi_0^*(r^N)(1-\exp\{i\int_0^t[V(\vec{r}_1+\vec{v}_k(t-t'),\vec{r}_2)-V(\vec{r}_1+\vec{v}_k t,\vec{r}_2)]\,dt'\})\Psi_0(\vec{r}_1+\vec{v}_k t,\vec{r}_2,\ldots,\vec{r}_N)\,d\tau^N . \tag{33}$$

Integrating over coordinates $\vec{r}_3,\ldots,\vec{r}_N$ introduces the two-particle density matrix, defined by

$$\rho_2(\vec{r}_1,\vec{r}_2;\vec{r}_1',\vec{r}_2')=N(N-1)\int\Psi_0^*(\vec{r}_1,\vec{r}_2,\vec{r}_3,\ldots,\vec{r}_N)\Psi_0(\vec{r}_1',\vec{r}_2',\vec{r}_3,\ldots,r_N)d\tau_3\cdots d\tau_N , \tag{34}$$

and yields for this term

$$\int\rho_2(\vec{r}_1,\vec{r}_2;\vec{r}_1+\vec{v}_k t,\vec{r}_2)(1-\exp\{i\int_0^t[V(\vec{r}_1+\vec{v}_k(t-t'),\vec{r}_2)-V(\vec{r}_1+\vec{v}_k t,\vec{r}_2)]\,dt'\})d\tau_1 d\tau_2 . \tag{35}$$

Putting $\vec{r}=\vec{r}_1-\vec{r}_2$ and again assuming translation invariance, this becomes

$$\Omega'\int\rho_2(\vec{r},0;\vec{r}+\vec{v}_k t,0)(1-\exp\{i\int_0^t[V(\vec{r}+\vec{v}_k(t-t'))-V(\vec{r}+\vec{v}_k t)]\,dt'\})d\tau . \tag{36}$$

Substituting Eqs. (31) and (36) into Eq. (28) yields the function $R(\vec{k},t)$ depending on the quantities anticipated above:

$$R(\vec{k},t)\cong\exp\frac{-\int\rho_2(\vec{r},0;\vec{r}+\vec{v}_k t,0)(1-\exp\{i\int_0^t[V(\vec{r}+\vec{v}_k(t-t'))-V(\vec{r}+\vec{v}_k t)]dt'\})d\tau}{\rho_1(0,\vec{v}_k t)}. \tag{37}$$

Equation (37) provides a useful approximation to the effect of target-atom interactions in altering the impulse approximation results for neutron scattering at high momentum and energy transfer. Two main approximations have been made to get to this result. The first consisted of the neglect of the time evolvement of the target-particle coordinates [induced by the operators $e^{\pm iH(t-t')}$ in Eq. (18)] while interacting with the struck particle (moving with a velocity v_k). The velocity v_k imparted to a target particle is much larger than a typical target-atom velocity, so that for

relatively soft collisions of target particles, this neglect seems relatively safe. For strong head-on collisions of the struck particle with other target particles, the readjustment of particle coordinates induced by the neglected operators $e^{\pm iH(t-t')}$ must play a large effect in preventing penetration into the hard-core region of the interaction, and here the approximation is dangerous. However, the situation encountered here is preferable to the one encountered in the expansion of the T product in Eq. (15). For example, if one were dealing with a Lennard-Jones potential, the quantity

$$\int_0^t U_j(v_k t')\,dt' = \int_0^t \sum_{\substack{m \\ m\neq j}} [V(\vec{r}_j - \vec{v}_k t', \vec{r}_m) - V(\vec{r}_j, \vec{r}_m)]\,dt'$$

in Eq. (15), and the equivalent quantity

$$\int_0^t [V(\vec{r} + \vec{v}_k(t-t')) - V(\vec{r} + \vec{v}_k t)]\,dt'$$

of Eq. (37), become undefined if the "trajectory" of the struck particle passes through the singularity of the potential. This divergence leads to an undefined expression for $S(\vec{k}, t)$ if evaluated from a finite number of terms from Eq. (15). In Eq. (37), the divergence occurs in the phase factor of an imaginary exponential and it therefore yields a well-defined result if some sensible limiting procedure is used. A tempting speculation is that the rapid oscillatory contributions from hard collisions will be small, mimicking the more physical picture in which the remaining particles will avoid close encounters through the action of $e^{\pm iH(t-t')}$.

The second approximation contained in the final expression for $R(\vec{k}, t)$ is concerned with truncating the cumulant expansion at the term ω_1 in Eq. (22). The neglected terms describe correlations between two or more passive target particles during their interaction with the struck target particle. Thus the approximate expression for $R(\vec{k}, t)$ contains multiple scatterings of the struck target particle by the remaining target particles, with each of the passive target atoms treated independently of each other. This approximation clearly requires that the correlation range between target atoms in the averaging state Ψ_0 be considerably larger than the interaction range for a pair of target particles. Although these conditions are not completely satisfied for relatively dense systems, inclusion of such "shadowing effects" seems inordinately difficult, requiring adding to $R(\vec{k}, t)$ terms involving three and higher-particle-density matrices. Since we envision experimental conditions under which corrections to the impulse approximation represented by $R(\vec{k}, t)$ are relatively small, the binary-collision approximation we have employed should provide a significant description of these final-state corrections.

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APPENDIX A

In this Appendix, we outline the derivation of the identity

$$T\exp[iH(t-t_0) + i\int_{t_0}^t U_j(v_k t')\,dt'] = T\exp[i\int_{t_0}^t e^{iH(t-t')} U_j(v_k t') e^{-iH(t-t')}\,dt'] e^{iH(t-t_0)}, \tag{A1}$$

which, for $t_0 = 0$ is Eq. (16) of the text. We choose to illustrate the equality by a direct iterative solution. More sophisticated proofs of this identity are possible [for example, by differentiating both sides of (A1) with respect to t_0 and rearranging].

Begin by defining the left-hand side of (A1) as $U(t-t_0)$:

$$U(t-t_0) = T\exp[iH(t-t_0) + i\int_{t_0}^t U_j(v_k t')\,dt']. \tag{A2}$$

Then we wish to show that $U(t-t_0)$ can be rewritten as the right-hand side of (A1). The time derivation of U is

$$\frac{dU(t-t_0)}{dt} = i[H + U_j(v_k t)]U(t-t_0). \tag{A3}$$

The equivalent integral equation for U is

$$U(t-t_0) = e^{iH(t-t_0)} + i\int_{t_0}^t e^{iH(t-t')} U_j(v_k t') U(t'-t_0)\,dt'. \tag{A4}$$

The iterated solution to (A4) is

$$\begin{aligned} U(t-t_0) = e^{iH(t-t_0)} &+ i\int_{t_0}^t e^{iH(t-t')} U_j(v_k t') e^{iH(t'-t_0)}\,dt' \\ &+ i^2 \int_{t_0}^t e^{iH(t-t')} U_j(v_k t')\,dt' \\ &\times \int_{t_0}^{t'} e^{iH(t'-t'')} U_j(v_k t'') e^{iH(t''-t_0)}\,dt'' + \cdots. \end{aligned} \tag{A5}$$

By manipulating the times appearing in the exponentials, one can factor out the operator $e^{iH(t-t_0)}$:

$$\begin{aligned} U(t-t_0) = [1 &+ i\int_{t_0}^t e^{iH(t-t')} U_j(v_k t') e^{-iH(t-t')}\,dt' \\ &+ i^2 \int_{t_0}^t e^{iH(t-t')} U_j(v_k t') e^{-iH(t-t')}\,dt' \\ &\times \int_{t_0}^{t'} dt''\, e^{iH(t-t'')} U_j(v_k t'') \\ &\times e^{-iH(t-t'')} + \cdots] e^{iH(t-t_0)}. \end{aligned} \tag{A6}$$

The factorization of $e^{iH(t-t_0)}$ is possible in all higher orders, and the terms in square brackets (A6) produce the time-ordered operator on the right-hand side of (A1):

$$U(t-t_0)=T\exp[i\int_{t_0}^{t}e^{iH(t-t')}U_j(v_kt')\times e^{-iH(t-t')}dt']e^{iH(t-t_0)}, \quad \text{(A7)}$$

which completes the demonstration.

APPENDIX B

In this Appendix an expansion is developed for a time-ordered operator which resembles the cumulant expansion of an exponential operator. The expansion will be applied to $S(\vec{k},t)$,

$$NS(\vec{k},t)=e^{i\omega_k t}\sum_{j,l}\langle e^{-i\vec{k}\cdot(\vec{r}_j-\vec{r}_l)}e^{i\vec{p}_j\cdot\vec{v}_k t}\times T\exp[i\int_0^t e^{iH(t-t')}U_j(v_kt')e^{-iH(t-t')}dt']\rangle, \quad \text{(B1)}$$

which may be rewritten

$$S(\vec{k},t)=e^{i\omega_k t}\left\langle\left(\sum_l e^{-i\vec{k}\cdot(\vec{r}_1-\vec{r}_l)}e^{i\vec{p}_1\cdot\vec{v}_k t}e^{iHt}\right)\times T\exp\left(i\int_0^t dt'e^{-iHt'}\sum_{m\neq 1}[V(\vec{r}_1-\vec{v}_kt',\vec{r}_m)-V(\vec{r}_1,\vec{r}_m)]e^{+iHt'}\right)e^{-iHt}\right\rangle. \quad \text{(B2)}$$

The technique will be to find $E(t)$, such that

$$e^{-E(t)}=\left\langle\theta_1\left[T\exp\left(i\int_{t_0}^{t}dt'\sum_m\theta_m(t')\right)\right]\theta_2\right\rangle, \quad \text{(B3)}$$

where $\theta_1,\sum_m\theta_m(t')$, and θ_2 are arbitrary operators later to be chosen so that Eq. (B3) can be applied to Eq. (B2).

Taking the logarithm of Eq. (B3) yields

$$E(t)=-\ln\left\langle\theta_1T\exp\left[i\int_{t_0}^{t}dt'\sum_m\theta_m(t')\right]\theta_2\right\rangle. \quad \text{(B4)}$$

Introduce a parameter λ and two operators $\Gamma_m(t)$ and $\theta_m(t,\lambda)$, such that

$$1-\Gamma_m=T\exp[i\int_{t_0}^{t}dt'\theta_m(t')], \quad \text{(B5)}$$

$$1-\lambda\Gamma_m=T\exp[i\int_{t_0}^{t}dt'\theta_m(t',\lambda)]. \quad \text{(B6)}$$

Note from Eqs. (B5) and (B6) that the partial time derivative of Eq. (B6) is

$$\lambda\theta_m(t)T\exp[i\int_{t_0}^{t}dt'\theta(t')]=\theta_m(t,\lambda)T\exp[i\int_{t_0}^{t}dt'\theta_m(t',\lambda)]. \quad \text{(B7)}$$

From the above three equations, the following properties of $\theta_m(t,\lambda)$ may be deduced:

$$\theta_m(t,0)=0, \quad \text{(B8)}$$

$$\theta_m(t,1)=\theta_m(t), \quad \text{(B9)}$$

$$\left.\frac{\partial\theta_m(t,\lambda)}{\partial\lambda}\right|_{\lambda=0}=\theta_m(t)T\exp[i\int_{t_0}^{t}dt'\theta_m(t')], \quad \text{(B10)}$$

$$\left.\frac{\partial^2\theta_m(t,\lambda)}{\partial\lambda^2}\right|_{\lambda=0}=2\theta_m(t)T\exp[i\int_{t_0}^{t}dt'\theta_m(t')]\times\{1-T\exp[i\int_{t_0}^{t}dt'\theta_m(t')]\}, \quad \text{(B11)}$$

etc.

Defining

$$E(t,\lambda)=-\ln\left\langle\theta_1T\exp\left(i\int_{t_0}^{t}dt'\sum_m\theta_m(t'm\lambda)\right)\theta_2\right\rangle, \quad \text{(B12)}$$

one can see from Eq. (B9) that $E(t)=E(t,1)$; and from Eqs. (B8) and (B12) that $E(t,0)=-\ln\langle\theta_1\theta_2\rangle$. Assume $E(t,\lambda)$ is analytic in λ in the unit circle. $E(t)$ may then be obtained by expanding $E(t,\lambda)$ in a Taylor's series about $\lambda=0$, evaluated at $\lambda=1$:

$$E(t)=E(t,0)+\left.\frac{\partial E(t,\lambda)}{\partial\lambda}\right|_{\lambda=0}+\frac{1}{2!}\left.\frac{\partial^2E(t,\lambda)}{\partial\lambda^2}\right|_{\lambda=0}+\cdots. \quad \text{(B13)}$$

With the use of Eqs. (B10) through (B12), the partial derivatives of $E(t,\lambda)$ with respect to λ, evaluated at $\lambda=0$, may be obtained; allowing one to rewrite Eq. (B13) as

$$-E(t)=\ln\langle\theta_1\theta_2\rangle-\sum_m\frac{\langle\theta_1\Gamma_m(t)\theta_2\rangle}{\langle\theta_1\theta_2\rangle}+\left\{\sum_{m_1\neq m_2}\left[\left\langle\theta_1\int_{t_0}^{t}dt_1\int_{t_0}^{t}dt_2\frac{\partial\Gamma_m(t_1)}{\partial t_1}\left(\frac{\partial\Gamma_{m_2}(t_2)}{\partial t_2}\right)\theta_2\right\rangle\Big/\langle\theta_1\theta_2\rangle\right]-\frac{\langle\theta_1\sum_m\Gamma_m(t)\theta_2\rangle^2}{2\langle\theta_1\theta_2\rangle^2}\right\}+\cdots, \quad \text{(B14)}$$

where the first three terms of the Taylor's series, Eq. (B13), are shown explicitly. The final result is

$$\left\langle\theta_1\left(T\exp\left[i\int_{t_0}^{t}dt'\sum_m\theta_m(t')\right]\right)\theta_2\right\rangle=\langle\theta_1\theta_2\rangle\exp(\omega_1+\omega_2+\cdots), \quad \text{(B15)}$$

$$\omega_1=-\sum\frac{\langle\theta_1\Gamma_m(t)\theta_2\rangle}{\langle\theta_1\theta_2\rangle},$$

$$\omega_2=\sum_{m_1\neq m_2}\left[\left\langle\theta_1\int_{t_0}^{t}dt_1\int_{t_0}^{t}dt_2\frac{\partial\Gamma_{m_1}(t_1)}{\partial t_1}\times\frac{\partial\Gamma_{m_2}(t_2)}{\partial t_2}\theta_2\right\rangle\Big/\langle\theta_1\theta_2\rangle\right]-\tfrac{1}{2}\left\langle\theta_1\sum_m\Gamma_m(t)\theta_2\right\rangle^2\Big/\langle\theta_1\theta_2\rangle^2. \quad \text{(B16)}$$

Identifying the θ_j operators to apply these results to (B2), we get the value for ω_1 quoted in Eq. (23) of the text.

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Influence of Vibrational, Rotational, and Reorientational Relaxation on Pulse Amplification in Molecular Amplifiers*

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Short-pulse amplification in gaseous molecular amplifiers is complicated by many aspects of atomic and molecular interactions. Among these are the various vibrational, rotational, and reorientational relaxational processes which influence the coupling of the energy stored in the molecular radiators to the electromagnetic field. The properties of plane-wave pulse amplification, especially in the saturated regime, are examined numerically in order to quantitatively determine the detailed effects of these relaxational phenomena. As expected, the results for saturated amplification show that the amount of extracted energy decreases significantly when the rotational relaxation time is sufficiently long in comparison to the pulse width. We also observe the development of pulse-shape variations which are a direct result of the collisional phenomena and differ qualitatively from the results obtained for amplifying media without an energy reservior. There is a tendency for the pulse lengths to increase owing to the energy transfer, in contrast to the strong narrowing effects which occur in the absence of the collisional processes. The influence of reorientational collisions is found to be small, accounting for less than a 20% effect on the over-all conclusions. Finally, we present results concerning the development of an asymptotic pulse shape in high-gain amplifiers. In this case, the pulse shape clearly exhibits the competition between the stimulated rate, which scales with the optical flux, and the collisional rates which are determined by the particle density. Calculations of this nature may be applied directly to CO_2, CO, and HF molecular amplifiers for both the electrically and chemically driven systems.

I. INTRODUCTION

Electromagnetic pulse amplification in gaseous molecular amplifiers is complicated by many aspects of atomic and molecular interactions. Among these are the multitude of relaxational processes which couple the molecular systems to one another as well as influence the interaction of the molecular radiators with the electromagnetic field. The perturbing fields which operate on the quantum-mechanical systems generally have a very complicated structure. There is, in addition to the coherent electric field $\vec{E}(t,z)$ of the amplified wave, a component due to collisions from neighboring particles which is rapidly varying in space, time, and direction. If the possibility of significant correlations arising from the presence of a coherent optical field is ignored,[1] then the perturbing field due to collisions can be regarded as a stochastic variable.[2] We do not take this approach here. Instead, we represent the relaxational processes by the appropriate phenomenological parameters. This choice arises quite naturally from the strong identification of the particular parameters with the corresponding physical mechanisms and relaxational processes. The dynamics of the coherent electric field $\vec{E}(t,z)$ are treated semiclassically in the customary way.[3]