# Transition between the Quasistatic and Impact Limits in Spectral Line Broadening

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A kinetic description of line broadening is used to express the conventional approximation schemes as limits of a more general point of view. A profile function is given in terms of a generalized collision rate. Central to the calculation of this rate is a perturbed phase function that accounts for possible strong coupling between perturbers and the particle coupled to the light. An approximate expression for this phase function is given which continuously describes the transition between the two limits.

## I. INTRODUCTION

To calculate a spectral line profile resulting from collisions, one usually employs a simplified model at the outset. The most common picture is characterized by two extreme limits for the speed with which a given perturbing collision may occur. In one limit, the collisions are regarded as instantaneous. Here one speaks of the impact approximation. The quasistatic approximation denotes the alternative limit in which the perturbers are taken as fixed in space according to some probability distribution.

In dense plasmas, for example, the impact approximation is applied to the electrons, whereas the effects of the ions are considered accurately described by the quasistatic approximation. This procedure is usually permissible for the central portion of the profile; however, as one moves away from the line center, its usefulness diminishes. Physically, one expects the core of the profile to be impact-dominated with the wings quasistatic, as shown in Fig. 1. To study the region of the profile bounded by the core and a wing, however, one must consider a more general picture.

The kinetic description of the absorber (emitter) distribution function $f$ is a convenient starting place. Knowledge of $f$ yields the induced atomic currents and thus the absorption (emission) line shape. Here the relative importance of the impact and quasistatic limits is revealed by the non-Markovian nature of the kinetic equation satisfied by $f$. In this paper, it is shown that the rate of change of $f$ to lowest order in the density of perturbers is

$$\frac{d}{dT}f(T)=\int_0^T d\tau C(\tau, T)f(T-\tau). \qquad (1.1)$$

It is observed that the two extreme limits emerge from (1.1), corresponding to approximate treatment of the collision integral. Further, if the translational motion of the absorber is ignored, the Laplace transform of (1.1) linearized in the external field is

$$f(\Omega+i\eta)=[\Delta\Omega+i\,\delta(\Omega+i\eta)]^{-1}f(0), \qquad (1.2)$$

defining the line-profile function. Here, $\Delta\Omega$ is the difference of the perturbed and unperturbed frequency, $f(0)$ is the initial distribution function, and $\sigma$ is the frequency-dependent width-shift function.

Equation (1.2) is important, since it is completely equivalent to (1.1) and thus, contains both extreme limits. The explicit expression for $\sigma$ demonstrates the role of *strong coupling* between the absorber and perturber, i.e., a fully non-Markovian solution is incompletely described by ordinary perturbation theory.

In Sec. II, the coupled equations of the one- and two-particle distribution functions are derived. In Sec. III, these equations are shown to yield a kinetic equation for $f$ in terms of the perturbed-phase solution for the absorber. The extreme limits are discussed within the framework of this kinetic equation in Sec. IV. Finally, an improved approximation scheme is introduced in Sec. V along with a brief comment on the relationship of the present theory to recent work in the theory of strong plasma turbulence.

## II. COUPLED EQUATIONS FOR $f$

The distribution function is directly related to the more general quantity

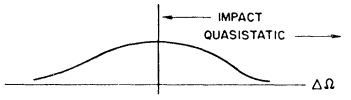


FIG. 1. Regions of profile where the two limits are expected to apply.

$$g^{<}(1,2)=(1/i)\langle\psi^{\dagger}(1)\psi(2)\rangle\equiv\langle\tilde{g}^{<}(1,2)\rangle\,. \tag{2.1}$$

The notation is chosen to coincide with that of Bezzerides.[1] $\psi^{\dagger}(1)$ and $\psi(2)$ create or destroy an absorbing atom at the space-time points $(\vec{x}_1, t_1)$ and $(\vec{x}_2, t_2)$, respectively. The internal labels for the atom are implicit in matrix notation. The bracket denotes averaging over some initial ensemble. As a function of the sum and difference coordinates, $X=\frac{1}{2}(x_1+x_2)$ and $x=x_1-x_2$, $\tilde{g}^{<}(X,x)$ satisfies

$$(i\partial_T+m^{-1}\partial_{\mathbf{X}}\cdot\partial_{\mathbf{x}})\tilde{g}^{<}(X,x)-[H_0,\tilde{g}^{<}(X,x)]$$

$$-[U(X+\tfrac{1}{2}x)\tilde{g}^{<}(X,x)-\tilde{g}^{<}(X,x)U(X-\tfrac{1}{2}x)]=0, \tag{2.2}$$

which is Eq. (5.13) of Ref. 1.

Equation (2.2) results directly from

$$(i\partial_{t_1}+1/2m\partial_1^{2})\tilde{g}^{<}(1,2)-[H_0+U(1)]\tilde{g}^{<}(1,2)=0, \tag{2.3}$$

and the corresponding equation for its $t_2$ time variation. $H_0$ is the Hamiltonian for the free stationary atom, and

$$U(1)=U^{\text{int}}(1)+U^{\text{ext}}(2)\,, \tag{2.4}$$

with $U^{\text{ext}}$ the interaction of the atom with an arbitrary external field.

The distribution function $f(\vec{\mathrm{X}},\vec{\mathrm{p}},\mathrm{t})$ is the ensemble average of

$$\tilde{f}(\vec{\mathrm{X}},\vec{\mathrm{p}},T)=\frac{i}{(2\pi)^3}\int d^3x e^{-i\vec{\mathrm{p}}\cdot\vec{\mathrm{x}}}\tilde{g}^{<}(X,x)\big|_{t=0}\,, \tag{2.5}$$

and from (2.2), $\tilde{f}$ satisfies

$$i(\partial_T+\vec{\mathrm{p}}/m\cdot\partial_X)\tilde{f}(\phi,T)-[H_0+U(X),\tilde{f}(\phi,T)]=0\,. \tag{2.6}$$

In (2.6), $(X,\vec{\mathrm{p}})$ are denoted by $\phi$. Terms due to acceleration of the c.m. of the absorbing atom are neglected in (2.6) since induced electric currents are assumed dominant. In the absence of internal structure, the second term of (2.6) vanishes, and the lowest-order acceleration term leads to the operator equivalent of the Vlasov equation for a plasma.

The apparent simplicity of (2.6) is deceptive since, for a plasma with dipole interaction between atom and perturbers,

$$U^{\text{int}}(X)=-\vec{\mathrm{m}}\cdot\vec{\mathrm{E}}(\vec{\mathrm{X}},T)$$

$$=-\vec{\mathrm{m}}\cdot\left(\sum_s\int d\phi'\vec{\epsilon}^{\,s}(\vec{\mathrm{X}},\vec{\mathrm{X}}')\tilde{f}_s(\phi',T)\right), \tag{2.7}$$

where $\vec{\epsilon}^{\,s}$ is the electric field produced by a single species $s$. Thus, in (2.6), $\tilde{f}$ is coupled to the full operator microfield. Only $f$ is needed, not $\tilde{f}$. However, taking the ensemble average of (2.6), one finds that $f$ is not self-determined. To solve the corresponding ensemble-averaged equation, one must know for all times,

$$f_s^{(1)}(\phi_1,\phi,T)=\langle\tilde{f}_s^{(1)}(\phi_1,\phi,T)\rangle$$

$$=\langle\tilde{f}_s(\phi_1,T)\;\tilde{f}(\phi,T)\rangle\,. \tag{2.8}$$

The correlation function $f_s^{(1)}$, in turn, is coupled to $f_{s,t}^{(2)}$, where

$$f_{s,t}^{(2)}(\phi_1,\phi_2,\phi,T)=\langle\tilde{f}_s(\phi_1,T)\tilde{f}_s(\phi_2,T)\tilde{f}(\phi,T)\rangle. \tag{2.9}$$

This procedure may be continued until the equivalent of the $N$-body Schrödinger equation is reached. To truncate this hierarchy of coupled equations, one usually assumes that a given $f^{(n)}$ may be expressed in terms of $f^{(n-1)}\times f$.

A rigorous justification for this truncation scheme requires an expansion of the solution for this hierarchy in terms of a power series in some small parameter. For the case of a gas consisting exclusively of neutral particles, this parameter is $\alpha=Nr_0^3$, where $N$ is the density of perturber and $r_0$ is the force range. As is familiar from kinetic theory, $\alpha$ may be thought of as being a measure of the probability that various numbers of particles will collide simultaneously. To lowest order in $\alpha$, one retains binary collisions and excludes three-particle collisional effects. Thus, to lowest order in density, three-particle correlations are ignored here.

For simplicity, the translational motion of all particles will be described classically. To obtain the classical limit, it is convenient to return to a fully spatial coordinate dependence for $f^{(2)}$. Thus, consider

$$\langle\psi^{\dagger}_s(\vec{1})\psi_s(\vec{1}')\tilde{f}(\phi)\psi^{\dagger}_t(\vec{2})\psi_t(\vec{2}')\rangle=\delta_{st}\delta^{(3)}(\vec{1}'-\vec{2})$$

$$\times\langle\psi^{\dagger}_s(\vec{1})\psi_s(\vec{2}')\tilde{f}\rangle+\langle\psi^{\dagger}_s(\vec{1})\psi^{\dagger}_t(\vec{2})\tilde{f}\psi_s(\vec{1}')\psi_t(\vec{2}')\rangle\,, \tag{2.10}$$

where the primed position coordinates are close to their corresponding unprimed coordinates. The second term on the right-hand side of (2.10) may be approximated by the sum of four terms. Two of these terms are negligible in the classical limit. The remaining two terms are

$$f_s^{(1)}(\phi_1,\phi,T)f_t(\phi_2,T)+f_s(\phi_1,T)f_t^{(1)}(\phi_2,\phi,T)\,, \tag{2.11}$$

where we have returned to the phase-space description.

With (2.10) and (2.11) the equation of motion for $f_s^{(1)}$ becomes

$$i(\partial_T+\vec{p}_1/m_s\cdot\partial_{\vec{X}_1}+\vec{p}/m\cdot\partial_{\vec{X}})f_s^{(1)}(\phi_1,\phi,T)$$
$$-[H_0+U^{\text{ext}},f_s^{(1)}(\phi_1,\phi,T)]+\vec{\epsilon}^s(\vec{X},\vec{X}_1)$$
$$\cdot[\vec{m},f_s^{(1)}(\phi_1,\phi,T)]$$
$$+\sum_t\int d\phi_2\vec{\epsilon}_t(\vec{X},\vec{X}_2)\cdot[\vec{m},f_t^{(1)}(\phi_2,\phi,T)]$$
$$\times f_s(\phi_1,T)=0. \tag{2.12}$$

Equation (2.12) is applicable to a homogeneous equilibrium plasma. Further, it has been assumed that the perturbing particles move free of interaction. The validity of this approximation as well as the classical path limit have been considered in the literature.[2] It should be noted that because of the special long-range characteristic of the interaction between an atom and a given charged particle of the plasma, the explicit integral expression for $\sigma$ must be cut off at approximately the Debye shielding distance.[3] The action of the commutators in (2.12) may be represented by a familiar notational device.[1] A dyad vector space is used to make the identification

$$\langle\beta|A_1B-BA_2|\alpha\rangle$$
$$\rightarrow\sum_{\alpha_1,\beta_1}\langle\alpha\beta|A_1{}^R-A_2{}^L|\alpha_1\beta_1\rangle\langle\alpha_1\beta_1|B\rangle, \tag{2.13}$$

where $A_1, A_2$, and $B$ are arbitrary matrix operators, and the new vector space is constructed in such a way that

$$\langle\alpha\beta|A_1{}^R|\alpha_1\beta_1\rangle\equiv\langle\alpha|A_1|\beta_1\rangle\delta_{\alpha\alpha_1}, \tag{2.14a}$$

$$\langle\alpha\beta|A_2{}^L|\alpha_1\beta_1\rangle\equiv\langle\alpha_1|A_2|\alpha\rangle\delta_{\beta\beta_1}, \tag{2.14b}$$

and $|B\rangle$ is defined by $\langle\alpha_1\beta_1|B\rangle\equiv\langle\beta_1|B|\alpha_1\rangle$. (2.15)

Since only commutators appear in (2.12), this formal equivalence may be written

$$[Q,f]\rightarrow L_Q|f\rangle, \tag{2.16}$$

where $L_Q$ is the tetradic operator corresponding to $Q$. With the use of this notation one writes, for the coupled set of equations for $f$ and $f^{(1)}$,

$$[i(\partial_T+\vec{p}/m\cdot\partial\vec{X})-(L_0+L_{\text{ext}})]|f\rangle$$
$$-\sum_s\int d\phi_2L_s(X,X_2)|f_s^{(1)}(\phi_2,\phi,T)\rangle=0, \tag{2.17}$$

and

$$[i(\partial_T+\vec{p}_1/m_s\cdot\partial_{\vec{X}_1}+\vec{p}/m\cdot\partial_{\vec{X}})$$
$$-(L_0+L_{\text{ext}}+L_s(\vec{X},\vec{X}_1))]|f_s^{(1)}(\phi_1,\phi,T)\rangle$$
$$-f_s\sum_t\int d\phi_2L_t(\vec{X},\vec{X}_2)|f_t^{(1)}(\phi_2,\phi,T)\rangle=0, \tag{2.18}$$

where $L_0$, $L_{\text{ext}}$, and $L_S$ are the tetradics corresponding to $H_0$, $U^{\text{ext}}$, and $-\vec{m}\cdot\vec{\epsilon}^s(X,X_1)$, respectively.

## III. KINETIC EQUATION FOR $f$

The simplest way to solve (2.17) and (2.18) is to consider their difference; thus, the quantity

$$|\delta f_s\rangle\equiv|f_s^{(1)}\rangle-f_s|f\rangle. \tag{3.1}$$

The quantity $|\delta f_s\rangle$ satisfies the equation

$$(i\partial_T+O_s)|\delta f_s(T)\rangle=L_sf_s|f(T)\rangle, \tag{3.2}$$

where $O_s$ in explicit coordinate representation is

$$O_s=(\vec{p}_1/m_s\cdot\partial_{\vec{X}_1}+\vec{p}/m\cdot\partial_{\vec{X}})$$
$$-[L_0+L_{\text{ext}}+L_s(\vec{X},\vec{X}_1)]. \tag{3.3}$$

Equation (3.2) is solved, given the solution of the corresponding homogeneous equation

$$(i\partial_T+O_s)U_s(T,T_0)=0, \tag{3.4}$$

with the boundary condition $U_s(T_0,T_0)=I$. Then it is a simple matter to show that

$$|\delta f_s(T)\rangle=U_s(T,T_0)|\delta f_s(T_0)\rangle$$
$$-i\int_{T_0}^{T}dT'U_s(T,T')L_sf_s|f(T')\rangle. \tag{3.5}$$

The propagation operator is easily determined from the homogeneous equation to be

$$U_s(T,T_0,\phi_1,\phi)=\exp[-i\int_0^{T-T_0}d\tau\,\mathfrak{L}_s$$
$$\times(\vec{X}(\tau),\vec{X}_1(\tau),T-\tau)]_-P(T-T_0). \tag{3.6}$$

The bracketed quantity is antitime ordered, and $\mathfrak{L}_s(\vec{X},\vec{X}_1,T)=L_0+L_{\text{ext}}(\vec{X},T)+L_s(\vec{X},\vec{X}_1)$. The

coordinates are shifted due to free flight, i.e., $\vec{X}(\tau)=\vec{X}-(\vec{p}/m)\tau$ and $\vec{X}_1(\tau)=\vec{X}_1-(\vec{p}/m_s)\tau$. The action of $P$ is to replace functions of $\vec{X}, \vec{X}_1$ by those translated through free flight over the time difference $T-T_0$.

The bracketed quantity in (3.6), ${U_s}^T(T-T_0)$, has a simple interpretation as the phase of the absorbing atom perturbed by the external field and by the presence of a single perturber particle. In the absence of any interaction this phase is $\exp[-iL_0(T-T_0)]$, which clearly agrees with ${U_s}^T(T-T_0)$. With (2.17) and (3.5), the kinetic equation for $|f\rangle$ is

$$[i(\partial_T+\vec{p}/m\cdot\partial_{\vec{X}})-(L_0+L_{\text{ext}})]|f(\phi,T)\rangle + i\int_0^T d\tau\,{\sigma}^T(\tau,\phi)|f(\phi(\tau),T-\tau)\rangle=0, \quad (3.7)$$

where $${\sigma}^T(\tau,\phi)=\sum_s {\sigma_s}^T(\tau,\phi) = \sum_s \int d\phi_1 f_s(\phi_1) L_s(\vec{X},\vec{X}_1) \times U_s(T,T-\tau) L_s(\vec{X},\vec{X}_1). \quad (3.8)$$

In (3.7), it has been assumed that the absorbing atom is initially uncorrelated with $T_0=0$.

## IV. IMPACT AND QUASISTATIC LIMITS

Limited descriptions of the collision rate $\sigma$ yield the conventional approximate schemes for the calculation of line profiles.

### A. Impact Limit

If $\sigma\exp(iTL_0)$ is peaked about $\tau=0$ with a width $\tau_{AC}$, one might write $|f(T-\tau)\rangle\simeq|f(T)\rangle$ in (3.7). (The free-phase factor removes the trivial time dependence of $|f\rangle$.) Further, if $T\gg\tau_{AC}$, the upper limit of the integral may be extended to infinity with small error. The result is a Markovian kinetic equation. Except for the possibility of overlapping lines, the corresponding line shape is Lorentzian. The $\tau_{AC}$ is a measure of the correlation time of the electric field of the plasma.

To consider the validity of this approximate procedure, take a definite matrix element of (3.7) for an isolated line. Expanding $\exp(i\tau L_0)f(T-\tau)$ about $\tau=0$ to terms linear in $\tau$, one must have

$${\tau_{AC}}^s\left[\frac{d}{dT}g(T)/g(T)\right]\ll 1 \quad (4.1)$$

to ignore non-Markovian corrections. The quantity $g(T)$ has the trivial oscillatory time variation of $f(T)$ removed. It is important to note that a certain portion of a line profile surrounding the line center can always be represented by a Lorentzian. The extent of this region according to (4.1) is ${\tau_{AC}}^s(\max)\Delta\omega\ll 1$, where $\Delta\omega$ measures the width of the Lorentzian, and ${T_{AC}}^s(\max)$ is correlation time for the species with the largest field correlation. Thus, the impact approximation is always valid in the core of a profile.

### B. Statistical Theory

To describe the wings of a profile, one needs accurate knowledge of the small $T$ dependence of $|f\rangle$. The peaking of $\sigma\exp(i\tau L_0)$ is then irrelevant, since the full extent of the integral is within ${\tau_{AC}}^s(\min)$. The simplest procedure is to ignore the dynamics of the perturbing field. This approximation is the basis of the quasistatic theory. Its validity is limited to that portion of the line set by

$${\tau_{AC}}^s(\min)\Delta\omega\gg 1. \quad (4.2)$$

Thus, the wings of a line are always quasistatic (statistical).

To see the connection between (3.7) and the usual form of the quasistatic theory it is useful to consider the second term of that equation as a single quantity – that is, to set

$$\sum_s\int_0^T d\tau\,{\sigma_s}^T(\tau,\phi)|f(\vec{X}(\tau),\vec{p},T-\tau)\rangle = \sum_s|Q_s(T,\phi)\rangle=|Q(T,\phi)\rangle. \quad (4.3)$$

The collision rate may be written

$${\sigma_s}^T(\tau,\phi)=\int d\phi_1 f_s(\phi_1)L_s(\vec{X},\vec{X}_1) \times\{i\frac{d}{d\tau}\bar{U}_s(T,T-\tau)-\bar{U}_s(T,T-\tau) \times[L_0+L_{\text{ext}}(\vec{X}(\tau),T-\tau]\}, \quad (4.4)$$

where $\bar{U}_s$ is $U_s$ except for $P$. Simple integration by parts gives

$$|Q_s(\tau,\phi)\rangle={I_s}^T(\phi,T)|f(\phi(T),0)\rangle - \int_0^T d\tau\,{I_s}^T(\phi,\tau)|Q(T-\tau,\phi(\tau))\rangle, \quad (4.5)$$

with $${I_s}^T(\phi,\tau)=i\int d\phi_1 f_s(\phi_1)L_s(\vec{X},\vec{X}_1)\bar{U}_s(T,T-\tau). \quad (4.6)$$

The equation for $|Q_s\rangle$ clearly converges for small $T$, indicating that this formulation of the collision term in (3.7) is relevant to the line wings. The kinetic equation for $|f\rangle$ with the lowest approximation for $|Q\rangle$ in the infinite mass limit is

$$[i\partial_T - (L_0 + L_{\text{ext}})]|f\rangle - \sum_s \int d\phi_1 f_s L_s \times (\vec{X}, \vec{X}_1)\overline{U}_s(T,0)\,|\,f(\phi,0)\rangle = 0. \quad (4.7)$$

The solution of (4.8) is

$$|f(\phi,T)\rangle = \sum_s \int d\phi_1 f_s(\phi_1)\overline{U}_s(T,0) \times |f(\phi,0)\rangle, \quad \text{where } m_s = \infty, \quad (4.8)$$

which, when linearized in the external field and Laplace-transformed, is equivalent to the asymptotic Holtsmark result.

Finally, the weak-coupling limit of (3.8), in which the unperturbed phase function is used for $\overline{U}_s$, yields the classical limit of the relaxation theory of line shapes.[3] Contrariwise, the above discussion should make clear in just what sense the proper description of the quasistatic theory is a strong-coupling limit.

## V. GENERAL LINE PROFILE

To obtain the line-profile function of the form expressed in (1.2), one must return to (3.7), the kinetic equation for $|f\rangle$. For spontaneous emission, $U^{\text{ext}} = 0$. (For absorption, the equation is linearized in the external field.) To simplify, take the absorbing atom as fixed at the origin (all line-shape calculations assume this at the outset). Then the Laplace transform of (3.7) gives

$$|f(\Omega + i\eta)\rangle = \int_0^\infty dT\, e^{i(\Omega + i\eta)T}|f(T)\rangle, \quad (5.1)$$

$$\text{or} \quad |f(\Omega + i\eta)\rangle = [\Omega - L_0 + i\sigma(\Omega + i\eta)]^{-1}\,|f(0)\rangle, \quad (5.2)$$

where $\sigma(\Omega + i\eta)$ is the transform of $\sigma$.

In the plasma, rapid transit of an electron results in a weak influence on the absorbing atom, allowing for a weak-coupling approximation, whereas a slowly moving ion has sufficient time to engage in strong interaction. In terms of $U_s(\tau)$, this approximate treatment is equivalent to

$$U_s(\tau) = e^{-i\tau L_0 P} \quad \text{(for electrons)}, \quad (5.3a)$$

$$= e^{-i\tau[L_0 + L_s(X, X_1)]} \quad \text{(for ions)}. \quad (5.3b)$$

It should be clear from the previous discussion that there exists a transition region in which this distinction according to species is inadequate for establishing which is the best limit, i.e., not all ions are quasistatic nor are all electrons impact.

Detailed calculation of $\overline{U}_s$ is difficult because of time ordering. For the limiting situations

$$\tau_c \ll \tau, \quad (5.4a)$$

$$\tau_c \gg \tau, \quad (5.4b)$$

where $\tau_c^{\,s} \equiv \dfrac{|\vec{X} - \vec{X}_1|}{|\vec{p}/m - \vec{p}_1/m_s|}$,

$$\overline{U}_s(\tau) \simeq \exp\left[-i\tau\left(L_0 + \frac{L_s(\vec{X}, \vec{X}_1)}{1 - \tau/\tau_c}\right)\right], \quad (5.5)$$

having taken the relative velocity parallel to the relative position vector. This approximation suggests that the distinction between the quasistatic and the weak-coupling limits should be made according to (5.4a) and (5.4b), respectively. In terms of $\sigma_p$, one would write

$$\sigma_p(\tau) \simeq \int_{\tau_c < \tau} d\phi f_s L_s (e^{-i\tau L_0} P) L_s + \int_{\tau_c > \tau} d\phi f_s L_s (e^{-i\tau(L_0 + L_s)}) L_s. \quad (5.6)$$

This simple cutoff procedure allows for a continuous transition between the two limits. In (5.6), the phase-space integration must be performed before determining $\sigma_p(\Omega^+)$ since the phase-space cutoffs are $\tau$-dependent. Detailed evaluation of these integrals is under study with the hope of obtaining an accurate description of that portion of the profile where the conventional scheme may need improvement.

The principal result of this paper is to demonstrate the nature of the quasistatic approximation from the point of view of an atom experiencing collisions with the particles of its surroundings. The simple picture of statistically shifted levels is replaced by a description in which the perturbing particles weakly scatter, but with an absorber (emitter) strongly perturbed in its phase. Thus one must introduce the perturbed phase function $\overline{U}_s(T)$ to replace the free-phase function $\exp(-iTL_0)$. In this regard it is interesting to note that Dupree,[4] in a theory of strong plasma turbulence, has reexamined the significance of particle-wave interactions in saturating certain plasma instabilities by introducing the concept of a perturbed particle orbit in place of the usual free motion for the scattering particle.[5] The use of a perturbed phase function $\overline{U}_s(T)$ has some parallel with the role of this perturbed-particle-orbit concept.

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# Brillouin Scattering in Simple Liquids: Argon and Neon

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Recent theoretical work has suggested that at high frequencies, there should be significant departure from classical hydrodynamic behavior in simple fluids. In particular, the frequency dependence of transport coefficients is no longer negligible and may introduce observable effects into the propagation of high-frequency sound. We have measured the sound velocity of high-frequency phonons (1–3 GHz) in liquid argon and liquid neon along their vapor-pressure equilibrium curves using the Brillouin scattering technique. The Brillouin spectra were excited with a single-mode argon-ion laser operating at 5145 or 4765 Å and were analyzed and detected with a Fabry-Perot interferometer and standard photoelectric techniques. Hypersonic (~3 GHz) velocities observed in argon decrease linearly from 850 m/sec at 85 °K to 742 m/sec at 100 °K and uniformly exhibit a small departure from low-frequency (1 MHz) data obtained under the same thermodynamic conditions. This effect is in qualitative agreement with theoretical-model predictions of a negative velocity dispersion at high frequencies. Our measurements of the sound velocity in liquid neon are the first in this material by any technique, and hence cannot be compared with ultrasonic values. The hypersonic velocity in neon decreases not quite linearly from 620 m/sec at 24.9 °K to 508 m/sec at 32 °K. When compared with results in other noble-gas liquids through corresponding-states arguments, these data suggest the existence of measurable quantum effects in the hypersonic velocity of liquid neon. In addition, an interesting change in slope of the velocity-versus-temperature curve (of 17%) is observed at 28 °K.

## I. INTRODUCTION

Although the technique of Brillouin scattering has been widely used to study very-high-frequency sound waves in liquids, scant information of this type is available for simple monatomic liquids.[1] Previous light scattering experiments on noble liquids have been concerned with sound-velocity behavior near temperatures of phase transition,[1] rather than with possible effects due to frequency dependence of the liquid's transport coefficients. Such information is difficult to obtain and yet is very important for further development of theories of the dynamics of the liquid state. Some recent theoretical work has indicated the possibility of nonclassical behavior for very-high-frequency sound waves (i.e., departure from the predictions of the Navier-Stokes equations). Gillis and Puff[2] have shown that for $\omega > \omega^*$ (where $\omega^*$ is some critical frequency in the liquid) sound waves propagate at some velocity intermediate between the adiabatic and the isothermal velocities, and that