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Nonequilibrium Excitation of Neutral Helium in Plasmas of Moderate Density

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Detailed rate equations for the excitation of neutral helium are solved, using a variety of assumed cross sections, and the resulting population densities are compared with measurements on stellarator Ohmic heating discharges and afterglows. It is shown that the experimental results can be satisfactorily reproduced by using for electron impact excitation cross sections for all optically allowed transitions a function $(\ln U)/U$ scaled according to the Bethe approximation, where U is the ratio of incident electron energy to the excitation energy. If the assumed cross sections differ from this near the threshold by more than a factor of $\frac{1}{3}$, satisfactory agreement with measurements is not obtained. Other items of interest in the analysis of laboratory and astronomical plasmas are also discussed.

I. INTRODUCTION

THE excitation and ionization of atomic systems by electron impact has been the subject of many investigations, both experimental and theoretical. Of necessity, however, direct measurements have been restricted almost exclusively to transitions involving ground states of neutral atoms and molecules. This, in turn, has led to almost as severe a limitation on theoretical studies because of the desirability of comparing calculated cross sections with experimental ones.

In the present investigation an indirect approach has been taken in an effort to obtain experimental information concerning collisionally induced transitions, especially among excited states of atoms. The method consists, essentially, of determining theoretical population densities of excited states by solving detailed rate equations, which include all collisional and radiative processes known to be of importance, and comparing them with population densities obtained by measurement. In the calculations, experimental cross sections are used when possible. For transitions for which experimental cross sections are not available or for which there is significant disagreement among different measurements, approximate theoretical cross sections are used. These are chosen to be as general as possible and incorporate a small number of parameters which are varied in an effort to improve agreement with measurements.

II. MEASUREMENTS

The measurements employed in the present work were made, for the most part, on helium discharges in the C stellarator, details of which have recently been published.[1,2] Electron densities were obtained from phase shifts of 4-mm microwaves and are believed to be accurate to within 15%. Concentrations of neutral atoms were deduced by measuring the pressure in the vacuum vessel before a stellarator pulse and correcting for the degree of ionization during the pulse. Electron temperatures were determined from electrical conductivity measurements for observations made during Ohmic heating. In the afterglow measurements of Hinnov and Hirschberg[3] and Hinnov,[4] which are also used in the present paper, temperatures were determined from the population densities of the higher excited states of neutral helium. Population densities of excited states in both Ohmic heating and afterglow discharges were obtained from absolute intensity measurements of the discharge spectra. The absolute intensity measurements for the stronger lines should be accurate to within 20%, with somewhat greater accuracy for relative intensities and poorer accuracy for weaker lines.

Results of the observations of Ohmic heating discharges and the afterglow measurements of Hinnov[4] are shown in Tables I and II. The lower excited states in the Ohmic heating discharges are considerably overpopulated with respect to Saha equilibrium, and in the afterglow discharges they are underpopulated.

TABLE I. Population densities of excited states of neutral helium in Ohmic heating discharges. (Superscripts denote powers of 10 to be applied.)

T_e (eV)	14.4	10.8	10.2	8.8	8.5	7.9	7.2	4.0
N_e (cm^{-3})	5.1^{12}	4.2^{12}	8.8^{12}	1.26^{13}	1.96^{13}	3.3^{13}	1.49^{13}	1.36^{13}
N_0 (cm^{-3})	4.1^{11}	8.8^{11}	1.44^{12}	4.7^{12}	6.3^{12}	1.8^{13}	8.5^{12}	8.2^{13}
State				N/g (cm^{-3})				
4^1S	9.3^{5}	6.6^{5}	1.51^{6}	1.61^{6}	2.2^{6}	4.8^{6}	1.48^{6}	3.6^{6}
5^1S	4.1^{5}	2.4^{5}	4.7^{5}	6.4^{5}	6.5^{5}	1.18^{6}	3.3^{5}	1.07^{6}
3^1P	1.11^{6}	6.9^{5}	2.5^{6}	4.8^{6}	7.4^{6}	1.91^{7}	4.5^{6}	1.74^{7}
4^1P	5.9^{5}	3.3^{5}	1.10^{6}	1.48^{6}	1.96^{6}	4.1^{6}	1.17^{6}	2.6^{6}
5^1P	3.2^{5}	1.24^{5}	4.7^{5}	5.6^{5}	5.0^{5}	1.09^{6}	4.8^{5}	6.6^{5}
3^1D	4.8^{5}	5.1^{5}	1.14^{6}	2.8^{6}	4.0^{6}	1.00^{7}	2.4^{6}	1.10^{7}
4^1D	4.1^{5}	3.0^{5}	7.9^{5}	1.24^{6}	1.45^{6}	3.0^{6}	9.1^{5}	2.6^{6}
5^1D	1.85^{5}	9.5^{4}	2.7^{5}	4.0^{5}	4.3^{5}	7.4^{5}	2.9^{5}	5.9^{5}
6^1D	8.0^{4}	4.5^{4}	1.37^{5}	1.32^{5}	1.40^{5}	2.8^{5}	8.0^{4}	2.3^{5}
7^1D	4.7^{4}	2.1^{4}	3.6^{4}	7.8^{4}	9.6^{4}	1.30^{5}	5.7^{4}	6.2^{4}
3^3S	2.3^{6}	2.7^{6}	6.3^{6}	1.23^{7}	1.19^{7}	2.6^{7}	1.06^{7}	1.96^{7}
4^3S	7.0^{5}	7.3^{5}	1.35^{6}	2.2^{6}	2.6^{6}	4.4^{6}	1.81^{6}	4.0^{6}
5^3S	1.76^{5}	1.86^{5}	3.1^{5}	4.6^{5}	5.8^{5}	9.7^{5}	4.2^{5}	7.7^{5}
3^3P	2.2^{6}	2.1^{6}	4.8^{6}	8.6^{6}	9.7^{6}	1.89^{7}	7.9^{6}	1.49^{7}
4^3P	5.0^{5}	4.9^{5}	1.12^{6}	1.62^{6}	1.82^{6}	3.4^{6}	1.40^{6}	2.4^{6}
5^3P	2.1^{5}	1.49^{5}	2.7^{5}	5.1^{5}	4.7^{5}	1.04^{6}	3.6^{5}	7.5^{5}
6^3P	6.6^{4}	3.9^{4}	7.7^{4}	1.61^{5}	1.40^{5}	2.3^{5}	1.05^{5}	2.2^{5}
3^3D	1.11^{6}	1.29^{6}	3.2^{6}	6.4^{6}	7.8^{6}	1.47^{7}	5.0^{6}	1.40^{7}
4^3D	3.6^{5}	3.3^{5}	7.9^{5}	1.35^{6}	1.53^{6}	2.9^{6}	1.13^{6}	2.6^{6}
5^3D	1.22^{5}	1.00^{5}	2.2^{5}	3.5^{5}	3.9^{5}	7.5^{5}	2.8^{5}	6.2^{5}
6^3D	3.7^{4}	3.0^{4}	7.3^{4}	1.14^{5}	1.05^{5}	1.97^{5}	9.6^{4}	1.72^{5}
7^3D	2.4^{4}	2.6^{4}	2.4^{4}	5.1^{4}	4.3^{4}	7.1^{4}	3.7^{4}	6.8^{4}

[1] R. M. Sinclair, S. Yoshikawa, W. L. Harries, K. M. Young, K. E. Weimer, and J. L. Johnson, Phys. Fluids **8**, 118 (1965).
[2] A. S. Bishop, A. Gibson, E. Hinnov, and F. W. Hofmann, Phys. Fluids **8**, 1541 (1965).
[3] E. Hinnov and J. G. Hirschberg, Phys. Rev. **125**, 795 (1962).
[4] E. Hinnov (unpublished).

TABLE II. Population densities of excited states of neutral helium in afterglow discharges.[a] (Superscripts denote powers of 10 to be applied.)

T_e (eV)	0.26	0.20	0.13
N_e (cm^{-3})	5.4^{12}	4.5^{12}	2.3^{12}
State		N/g (cm^{-3})	
4^1S	1.18^5	1.67^5	1.19^5
5^1S	1.17^5	1.75^5	1.26^5
3^1P	2.6^4	4.0^4	2.9^4
4^1P	8.8^4	1.08^5	6.9^4
5^1P	1.09^5	1.45^5	1.18^5
3^1D	4.0^4	5.2^4	3.7^4
4^1D	7.0^4	1.00^5	6.3^4
5^1D	7.6^4	1.18^5	9.3^4
4^3S	1.30^5	1.93^5	1.26^5
5^3S	1.36^5	1.96^5	1.60^5
3^3P	6.6^4	9.8^4	8.4^4
4^3P	1.47^5	2.01^5	1.39^5
5^3P	1.20^5	1.82^5	1.46^5
3^3D	6.3^4	8.7^4	5.1^4
4^3D	1.03^5	1.48^5	8.6^4
5^3D	9.7^4	1.51^5	1.15^5

[a] Reference 4.

III. CALCULATIONS

For each discharge condition for which measurements were available, population densities were calculated using essentially the same procedure as was used by Bates, Kingston, and McWhirter[5] to obtain collisional-radiative recombination rate coefficients for hydrogenic ions. The rate equation of the pth state is written as

$$\frac{dN(p)}{dt}=-N(p)\{N_e[S_e(p,c)+\sum_{q\neq p} S(p,q)] +\sum_{q<p} A(p,q)\}+N_e\sum_{q\neq p} N(q)S(q,p) +\sum_{q>p} N(q)A(q,p)+N^+N_e[S_e(c,p)+S_\nu(c,p)], \quad (1)$$

where $N(p)$ is the concentration of atoms in state p, N_e is the concentration of electrons, N^+ is the concentration of singly charged ions, $S_e(p,c)$ is the rate coefficient for ionization by electron impact, $S(p,q)$ is the rate coefficient for excitation from state p to state q by electron impact, $A(p,q)$ is the Einstein transition probability for a spontaneous radiative transition from state p to state q, and $S_\nu(c,p)$ is the rate coefficient for two-body radiative recombination to state p. In the sums $q<p$ is taken to mean $E_p<E_q$, where E_p is the energy required to ionize an atom in state p. $S(q,p)$ and $S_e(c,p)$ are obtained from $S(p,q)$ and $S_e(p,c)$, respectively, by detailed balance.

The set of equations represented by Eq. (1) can be reduced to a manageable number by assuming that all states above a certain level are in Saha equilibrium with the free electrons. In the present calculations it was assumed that all states with a principal quantum number greater than 20 were populated according to Saha's equation,[6]

$$N(q)=\frac{N_eN^+}{2g^+}g_q\left(\frac{2\pi\hbar^2}{mkT_e}\right)^{3/2}\exp\left(\frac{E_q}{kT_e}\right), \quad (2)$$

where g_q and g^+ are the statistical weights and $N(q)$ and N^+ are the concentrations for state q and the ground state of the ion.

An infinite sum in Eq. (1), such as $\sum_{q\neq p} N(q)S(q,p)$, converges rapidly and can safely be truncated when a given term is small compared to the sum up to that term.

Under all circumstances in the present work the time derivative in each rate equation, except for that of the ground state, can be neglected in comparison with other terms in the equation. One additional equation arises from the requirement of conservation of particles. There results a set of $n+1$ simultaneous linear equations from which the concentrations of n states and the time derivative of the ground-state population can be found.

In order to be able to deduce as much as possible from a detailed comparison of calculated and measured excited state concentrations, each state for which measurements could be obtained was treated as a separate state in the calculations. For most of the Ohmic heating discharges, populations of ^1D and ^3D states could be measured fairly reliably up to states with a principal quantum number of 7. Therefore, S, P, and D states with principal quantum numbers less than 8 were treated as separate states in the calculations. Corresponding states with a given principal quantum number and multiplicity but higher orbital angular momenta were lumped together and assumed to be hydrogenic. Similarly, all states with principal quantum numbers from 8 to 20 were taken to be hydrogenic. Thus 35 singlet and 34 triplet states were treated as distinct in the calculations.

A detailed description of the transition probabilities and cross sections used in the calculations and a more complete discussion of results are given in Ref. 7.

IV. RESULTS

1. Afterglow Discharges

The average electron energies in typical afterglow discharges are of the same order as transition energies among the higher excited atomic states, making it

[5] D. R. Bates, A. E. Kingston, and R. W. P. McWhirter, Proc. Roy. Soc. (London) **A267**, 297 (1962).

[6] This is justified by the fact that collisional rates become so large for transitions among highly excited states that Saha equilibrium is assured for states above some high level. The choice of $n=20$ as the quantum number of the state above which Saha equilibrium was assumed was dictated by the limits of a table of hydrogen oscillator strengths used in the calculations. If calculations of population densities are made for Ohmic heating discharges assuming Saha equilibrium for all states above $n=10$, the results differ from those for an $n=20$ cutoff by about 70% at the $n=10$ level and by a negligible amount for $n\leq 5$.

[7] L. C. Johnson, Princeton Plasma Physics Laboratory Report No. MATT-436 1966 (unpublished).

possible for an indirect approach of the type under consideration to yield quantitative information about collision cross sections for these transitions. The principal theoretical results which can be tested are the cross sections of Gryziński,[8,9] based upon classical theory, and those of Saraph,[10] based upon the impact-parameter approximation of Seaton.[11] Calculations[5] based upon the Gryziński cross sections are known to be in agreement with experiment,[3,12] but the cross sections of Saraph are considerably smaller near the threshold than those of Gryziński (by about a factor of $\frac{1}{10}$ for $n=15\rightarrow n=16$ transitions in hydrogen), and it was not apparent that the smaller cross sections might not also be consistent with the measurements.

To test these theoretical cross sections, collisional-radiative recombination rate coefficients were calculated for several afterglow discharges. Cross sections for optically allowed transitions were assumed to be of the form

$$\sigma(p,q)=\sigma_0(p,q)F_{pq}(U_{pq}), \tag{3}$$

where

$$\sigma_0(p,q)=4(E_{\mathrm{H}}/E_{pq})^2 f_{pq}\pi a_0^2 \tag{4}$$

and

$$F_{pq}(U_{pq})=\left\{1-\exp\left[-\frac{E_{pq}}{A}(U_{pq}+1)\right]\right\}\frac{\ln U_{pq}}{U_{pq}}. \tag{5}$$

In these expressions E_{pq} is the transition energy from state p to state q, f_{pq} is the absorption oscillator strength, U_{pq} is the ratio of incident electron energy to excitation energy,

$$U_{pq}=E/E_{pq}, \tag{6}$$

E_{H} is the ionization energy of hydrogen, and a_0 is the first Bohr radius. Equations (3), (4), and (5) reduce to the Bethe approximation[13] at high energies and satisfactorily reproduce the Saraph cross sections when the parameter A has the value 3.4 eV. When A approaches zero, giving a $(\ln U_{pq})/U_{pq}$ shape for all transitions and all energies of incident electrons, they are more nearly in agreement with the Gryziński cross sections.

Typical results are shown in Fig. 1. Here the solid curves represent the ratios of calculated-to-measured rate coefficients plotted as functions of the logarithm of the adjustable parameter A for several conditions. The curves are labeled with the electron temperatures, in eV, and start on the right with A chosen to agree with Saraph's results.

The calculated recombination rate coefficients are clearly too small when Saraph's results are used, and the discrepancy increases as the electron temperature is decreased. When the parameter A approaches zero, so that $F_{pq}(U_{pq})$ in Eq. (5) approaches $(\ln U_{pq})/U_{pq}$ for all optically allowed transitions, the calculated rates are more nearly equal to the measured rates and no longer obviously correlated with temperature.

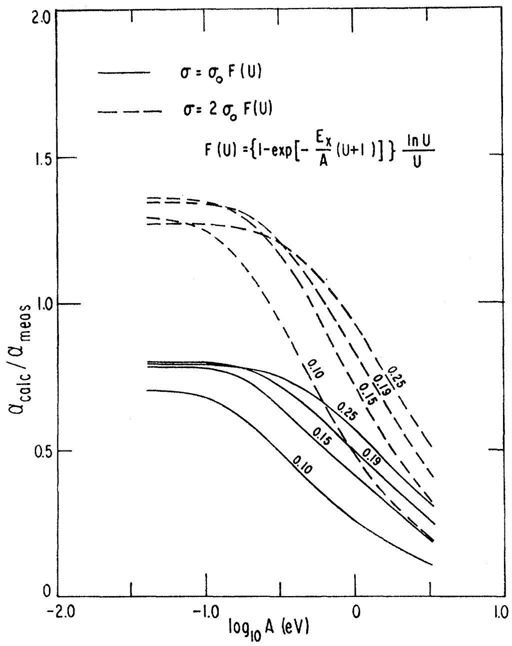

FIG. 1. Ratios of calculated to measured collisional-radiative recombination rate coefficients for helium afterglows.

As a further illustration of the sensitivity of such calculations to moderate changes in the assumed cross sections, the dashed curves in Fig. 1 show similar results obtained by doubling all collisional cross sections. The calculated rate coefficients in the $(\ln U)/U$ limit are seen to increase by about 70% when the cross sections are doubled. Although the spread of values for different temperatures does not change appreciably in the $(\ln U)/U$ limit, it is magnified in the other extreme. This clearly shows that a reduction in the cross-section shape near threshold which depends strongly upon excitation energy must be incorrect, at least for average $n\rightarrow n+1$ cross sections.

Because the collisional-radiative recombination rate coefficient is dominated by the contributions from one or two atomic states, which probably accounts for the fact that the curves of Fig. 1 are not asymptotic to unity, detailed comparisons of calculated and measured population densities were made for all states for which measurements were available. A number of rather general expressions were tried for $F_{pq}(U_{pq})$ in Eq. (3). The most favorable comparisons were obtained for

$$F_{pq}(U_{pq})=\{1-\exp[-0.2(U_{pq}+1)]\}(\ln U_{pq})/U_{pq}. \tag{7}$$

Although such a general formula cannot be expected to be correct for each possible transition, it should represent a reasonably accurate average cross-section shape for optically allowed transitions among helium states with principal quantum numbers greater than 2.

It should perhaps be emphasized that a $(\ln U)/U$

[8] M. Gryziński, Phys. Rev. **115**, 374 (1959).
[9] M. Gryziński, Phys. Rev. **138**, A336 (1965).
[10] H. E. Saraph, Proc. Phys. Soc. (London) **83**, 763 (1964).
[11] M. J. Seaton, Proc. Phys. Soc. (London) **79**, 1105 (1962).
[12] E. Hinnov, Phys. Rev. **147**, 197 (1966).
[13] H. A. Bethe, Ann. Phys. (Leipzig) **5**, 325 (1930).

shape, scaled according to the Bethe approximation, is expected to be valid for allowed transitions among excited states at sufficiently high energies of incident electrons ($U \gg 1$). The point of interest here is that the actual cross sections must not differ from this asymptotic relationship by more than a factor of about $\frac{1}{3}$ even at small values of U.

2. Ohmic Heating Discharges

Before discussing the conclusions to be drawn from a study of Ohmic heating discharges, it is of interest to consider some examples of the sensitivity of the calculated population densities to moderate changes in the assumptions. Figures 2 and 3 show ratios of calculated-to-measured population densities for the 8.8-eV discharge of Table I. For condition (a) a $(\ln U)/U$ shape is assumed for all optically allowed transitions, and experimental cross sections are used for forbidden transitions from the ground state. The other conditions, designated as (b) through (h), differ from (a) as follows:

(b) $F_{pq} = \{1 - \exp[-0.2(U_{pq}+1)]\}(\ln U_{pq})/U_{pq}$ for all allowed transitions.

(c) All collisional rates are doubled for transitions not involving the ground state.

(d) Collisional rates for $n=2 \rightarrow n=3$ transitions are reduced by one-half.

(e) Optically forbidden collisional transitions from the ground state to states with $n>2$ are omitted entirely.

(f) All optically forbidden transitions from the ground state are reduced by one-half.

(g) The concentration of neutral atoms is doubled.

(h) The concentration of electrons is doubled.

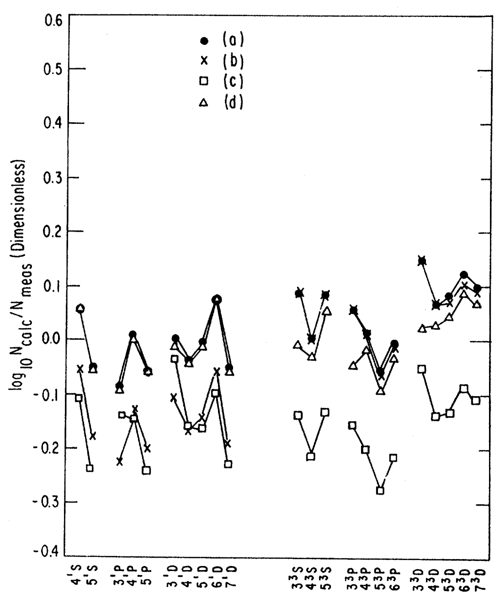

FIG. 2. Ratios of calculated to measured population densities for an Ohmic heating discharge for assumed conditions (a), (b), (c), and (d) described in the text.

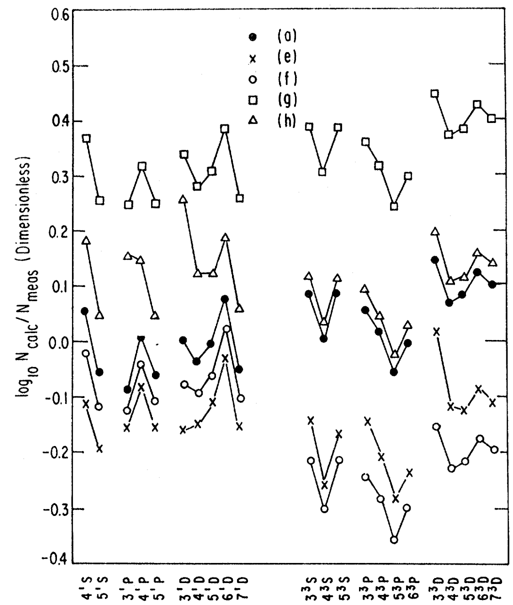

FIG. 3. Ratios of calculated to measured population densities for an Ohmic heating discharge for assumed conditions (a), (e), (f), (g), and (h) described in the text.

From Figs. 2 and 3 a number of conclusions can be drawn, among which are the following:

(1) The large population densities of excited states in Ohmic heating discharges, with respect to Saha equilibrium (a difference of about a factor 10^4 for the lower triplet states in this example), can be quantitatively accounted for by collisional and radiative transitions. The triplet states of neutral helium in such discharges are populated primarily by forbidden transitions from the ground state to the $n=2$ triplet states followed by excitation to higher states through allowed transitions, and not by direct $1 \rightarrow n>2$ transitions.

(2) The conclusions drawn from the study of afterglow discharges, namely, that Eqs. (3), (4), and (7) give reliable cross sections for allowed transitions among the higher excited states, do not lead to contradictions when applied to Ohmic heating discharges. For transitions involving at least one state with a principal quantum number less than 3, however, some modification is required.

(3) Of the examples shown, only assumption (d), i.e., the reduction of collisional rates for $n=2 \rightarrow n=3$ transitions, while other transition rates are left unchanged, removes the calculated overpopulation of the $n=3$ triplet states relative to higher states. This calculated overpopulation was found in all of the Ohmic heating discharges of Table I. An assumption such as (d) is the only simple assumption which could be found to remedy the situation without contradicting other results. In any event, the situation can be clarified when it is possible to make additional measurements, including measurements of 2^3P population densities,

which should depend rather sensitively upon the $2 \to 3$ rate coefficients.

(4) The calculated population densities of singlet states are sensitive to moderate changes in the assumed cross sections for $1^1S \to n^1P$ transitions. Analysis of the data available indicates that the cross-section shape observed by Thieme,[14] normalized to the Born approximation calculation of Seaton,[15] is as accurate for the $1^1S \to 3^1P$ transition as any in the literature, and that, in particular, it appears to be preferable near threshold to the measurements of St. John, Miller, and Lin.[16] Reliable cross sections for other resonance transitions can be obtained by scaling according to Eq. (4).

The conclusions mentioned are by no means all that can be drawn from an investigation such as the present one. These, however, are the ones of interest which are least likely to be affected by errors in measurements and by entrapment of resonance radiation. Further results, as well as confirmation of Eq. (7) and points (3) and (4) above, must await additional measurements.

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[15] M. J. Seaton, in *Atomic and Molecular Processes*, edited by D. R. Bates (Academic Press Inc., New York, 1962), Chap. 11.
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Convergent Kinetic Equation for a Classical Plasma*

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A general quantum-mechanical transport equation is used to derive a kinetic equation for an electron gas which in the classical limit is not subject to the usual short-range divergence and is exact to first order in the plasma parameter. The method is based on a direct analogy with the well-known equilibrium theory of the electron gas. No arbitrary separations or cutoffs are necessary. The resulting collision integral is similar to that of Weinstock and of Frieman and Book, but the Boltzmann and Fokker-Planck terms are evaluated for the static screened Coulomb potential instead of the bare Coulomb potential. It is shown that the equation of Guernsey, although convergent, does not contain all first-order contributions in the plasma parameter, and that the equations of Weinstock and of Frieman and Book must be carefully interpreted to achieve correct results. Numerical results, given in the classical limit for the dc electrical conductivity, explicitly exhibit the dominant and nondominant terms.

I. INTRODUCTION

THE derivation of a convergent kinetic equation for a classical plasma has been the subject of much attention recently.[1–5] As is well known, the usual kinetic equation for a classical plasma,[6–8] commonly referred to as the Lenard-Balescu equation, is logarithmically divergent for small impact parameters because of the neglect of close collisions. The problem can be made more specific by focusing our attention on a particular transport quantity such as the dc electrical conductivity. The model considered is an electrically neutral system of electrons and infinitely massive ions. The static conductivity σ can be written as

$$4\pi\sigma = \omega_p^2/\Gamma\,, \tag{1}$$

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