The |R| dependence in Eq. (11b) was introduced phenomenologically, and we only have to verify the angular dependence. But if the induced moment lies along the radius vector, then we must have

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PHYSICAL REVIEW A

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# Behavior of $He(2^{3}S)$ Metastable Atoms in Weakly Ionized Helium Plasmas\*

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Investigations concerning the behavior of the He( $2^{3}S$ ) population in active-discharge and afterglow helium plasmas are described. Good quantitative agreement is obtained between measured He( $2^{3}S$ ) densities in active discharges and predicted values based on the solution of a nonlinear rate equation. The rate coefficient for ionizing triplet-metastable-metastable collisions is measured in afterglow helium plasmas for the pressure range 10-40 Torr; close agreement is obtained with the value reported by Phelps and Molnar, though consideration of effects due to electrons suggests a downward revision of about 20%. The electron temperature decay in the afterglow is measured and found to agree closely with the decay expected on the basis of calculated energy source and loss processes which affect the electron gas. In this work, measurement of the He( $2^{3}S$ ) metastable density is accomplished by the novel use of a He-Ne laser operating at 1.0798  $\mu$  in an interferometer as well as by the conventional optical-absorption method. Advantages and disadvantages of this interferometer technique are discussed.

## I. INTRODUCTION

The behavior of atoms excited to the  $2^{3}S$  metastable state of helium has been studied in detail by many previous authors.<sup>1-7</sup> In this paper we report further investigations concerning the tripletmetastable populations of weakly ionized activedischarge and afterglow helium plasmas. In our work we attempt to describe accurately the steadystate triplet populations in active discharges in pure helium, the electron temperature decay in helium afterglows, and we perform a remeasurement of the triplet-metastable-metastable collision rate coefficient. Our experimental apparatus includes a laser interferometer operating in the infrared which is used for  $2^{3}S$  metastable density

measurements, as described in Sec. II. Tripletmetastable concentrations are inferred from plasma refractivity measurements by use of the standard formula for anomalous dispersion.<sup>8</sup> In Sec. III, a continuity equation describing the production and loss of triplet-metastable atoms in active discharges in pure helium is presented. The equation uses cross sections and calculated electron-energy distribution functions which are in the literature. Good quantitative agreement is obtained in the range of pressures and currents studied between the measured triplet-metastable densities and the densities predicted by the solution to the continuity equation. In Sec. IV, the electron temperature decay of an afterglow helium plasma is considered. We show that, with knowledge of triplet-metastable

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and electron densities, the electron temperature decay can be predicted quite accurately from an electron-energy balance equation. And in Sec. V, a remeasurement of the rate coefficient for ionizing collisions between triplet-metastable states is described. From a simple analysis of our data, one obtains a rate coefficient which is almost identical to the value reported previously by Phelps and Molnar.<sup>2</sup> However, calculations and experimental results indicate that some of the measured loss rate is due to superelastic electron-metastable collisions, and suggest as much as a 20% downward revision in the metastable-metastable collision rate coefficient.

## **II. EXPERIMENTAL APPARATUS AND TECHNIQUES**

The plasmas studied in this experiment were contained in a cylindrical quartz discharge tube, 46 cm long by 1-cm diam. The ends of the tube were closed by quartz flats placed at Brewster's angle in order to permit the use of laser diagnostics. Research-grade helium gas was used; it was cataphoretically cleaned for several hours prior to afterglow measurements at 10 Torr and above. The intensity of neon impurity lines in the afterglow was reduced below the threshold of detectability by the cataphoresis cleaning; this represented a reduction of more than two orders of magnitude in the neon impurity concentration. While impurities still may have affected recombination, it is estimated that they were a negligible factor in the destruction of metastables in the active-discharge and afterglow plasmas studied.

Plasmas for our afterglow studies were produced by current pulses which had insufficient energy to appreciably affect the mean gas temperature. However, neutral gas heating was an important effect in the active discharges studied. In this case, the discharge-tube wall temperature was measured with a thermocouple and the energy input to the gas was calculated from the discharge current and the longitudinal electric field. With this information and the measured pressure, the neutral gas density was calculated by solving the heat equation, as has been done elsewhere. $^{9, 10}$  It was found that the mean gas density averaged across the diameter of our discharge tube was lowered 30% by currents of about 100 mA at pressures near 1 Torr, when the gas pressure was held constant. Thus modest current densities were found to have significant effects on neutral gas densities.

In active discharges, the longitudinal electric field was calculated from the potential drop measured between two tungsten probes placed in an auxiliary discharge tube operating under the same conditions as the main tube. This electric field was used in the aforementioned neutral gas heating calculations and in calculations of E/p', the ratio

of electric field to gas pressure (reduced to 300 °K), which was used to characterize the electronenergy distribution functions in our active discharges.

Electron densities in both active-discharge and afterglow plasmas were calculated from the measured resonant frequency shifts of a C-band  $TM_{010}$ microwave cavity which surrounded a portion of the discharge tube. Use was made of a computer program<sup>11</sup> which solved the wave equation for the geometry employed, taking into account the changes in the field distributions due to the glass tube and the plasma. Electron temperatures in decaying plasmas were inferred from measurements of electron collision frequency for momentum transfer made with the same cavity. The cavity response to fixed frequency excitation was monitored as the plasma decayed. The cavity Q as measured from the response curve was related to the collision frequency with the following formula:

$$\langle \nu \rangle = \frac{\omega_a \,\omega_b}{2 \,|\, \Delta \omega \,|} \left[ \frac{1}{Q_b} - \frac{1}{Q_a} \left( \frac{2\omega_b}{\omega_a} - 1 \right) \right] \,, \tag{1}$$

where  $\langle \nu \rangle$  is the electron collision frequency for momentum transfer averaged over the electronenergy distribution function,  $\omega_a, \omega_b$  is the cavity resonant frequency without/with the plasma,  $Q_a$ ,  $Q_b$  is the cavity Q without/with the plasma, and  $\Delta \omega = \omega_h - \omega_a$ . This formula may be obtained from elementary analysis (see the Appendix) and entails the approximations that  $\omega_a$  and  $\omega_b$  are comparable,  $Q_a$  is considerably greater than  $Q_b$ ,  $\langle \nu \rangle$  is spatially uniform, and  $(\langle \nu \rangle / \omega_a)^2 \ll 1$ . The ratio of real to imaginary parts of the plasma dielectric constant has been taken as  $\langle \nu \rangle / \omega_b$ , which is valid under many conditions.<sup>12</sup> The average electron-neutral collision cross section for momentum transfer<sup>13</sup> was taken to be  $5.6 \times 10^{-16}$  cm<sup>2</sup> for 300 °K electrons and 5.8×10<sup>-16</sup> cm<sup>2</sup> for 600  $^{\circ}$ K electrons. The contribution to  $\langle \nu \rangle$  due to electron-ion collisions was small (10%) according to calculations based on Spitzer's work<sup>14</sup>; its effect was taken into account by including an average spatially uniform correction term in our calculations. The formulation of cavity perturbation theory used here is advantageous for determining collision frequencies in helium because it should be slightly more accurate than the standard approach in which  $\omega_a \omega_b$  is replaced by  $\omega_a^2$ , etc. The increased accuracy of this formulation is not of great importance in this work, however, because of the use of other, more sizeable approximations.

Triplet-metastable density measurements were performed by use of both interferometric and absorption techniques. The experimental arrangement is shown in Fig. 1. As can be seen, the endlight from a discharge tube was used for absorption measurements; corrections for the ex-



FIG. 1. Experimental apparatus.

pected self-absorbed emission line shape were made by performing multiple-path absorption measurements.<sup>8</sup> High accuracy was not required here since all accurate absolute densities and spatial distributions were measured with the laser interferometer. The laser was operated on the 1.0798- $\mu$  transition of neon<sup>15</sup> (2s<sub>3</sub> - 2 $p_7$ ) which is 32 Å away from the 1.0830- $\mu$  absorptive transitions from the  $He(2^{3}S)$  level. The interferometer was a standard type operated in the s = 8 configuration as described in Ref. 16. The line shape of the 1.0830- $\mu$  transition in the remote wings at 1.0798  $\mu$  was assumed to be given by a Lorentzian profile with a damping parameter determined by the radiative transition probability.<sup>17</sup> The anomalous dispersion<sup>8</sup> at the laser wavelength due to atoms in the triplet-metastable level was thus calculated to give one fringe per change of  $7 \times 10^{12}$ metastable atoms cm<sup>-3</sup>. Accurate measurement of fractional fringe shifts ( $\approx 0.1$  fringe) was facilitated by translating the external mirror during measurements, <sup>18</sup> and thus small changes in the resulting repetitive fringe pattern could be detected easily. Refractive effects caused by changes in neutral densities were significant in some cases; operation of the interferometer at several wavelengths allowed these effects to be determined.<sup>16</sup>

This interferometric technique for metastable density measurement is advantageous because the measured quantity is linearly proportional to metastable density; this is not the case with optical-absorption measurements. This technique is also quite accurate, the usual limitations being in knowledge of A coefficients, <sup>17</sup> wavelengths, <sup>17</sup> and easily measured geometrical lengths. (In our work, the largest source of error probably was due to the assumed longitudinal invariance of the plasma.) This technique also allows highly resolved spatial measurements to be made, the laser beam diameter being the limiting factor. However, the temporal coherence of the 1.0798- $\mu$ laser line was found to be much worse than that of the 0.6328-, 1.1523-, and 1.1767- $\mu$  lines, and this was a serious drawback to the technique. We attribute this frequency instability to small fluctuations in the helium triplet-metastable population inside the laser tube itself. This would then be similar to the situation observed previously at 0.6328  $\mu$ .<sup>18,19</sup> Operation of the laser at 1.0844  $\mu$ would improve the threshold sensitivity of our system as would a switch to laser heterodyning, but these changes would be accompanied by greater instability problems which would have to be overcome by synchronous detection schemes. This instability problem could be entirely eliminated, of course, if oscillation at 1.0798 or 1.0844  $\mu$  could be achieved in pure neon, as has been done at 1.1523  $\mu$ .<sup>20</sup>

## **III. ACTIVE DISCHARGE INVESTIGATION**

In this section we consider triplet-metastable atom behavior in a dc-excited active discharge in helium. The following continuity equation is taken to be descriptive of the production and loss of triplet metastable atoms:

$$\frac{\partial M}{\partial t} = D\nabla^2 M + \alpha_1 n_e N - \alpha_2 n_e M - \alpha_3 M^2 = 0 , \qquad (2)$$

where M is the He( $2^{3}S$ ) density, N is the groundstate helium-atom density,  $n_e$  is the electron density, D is the triplet-metastable diffusion coefficient, <sup>3</sup>  $\alpha_1$  is the rate coefficient for production of triplet atoms by electronic collisions with groundstate helium atoms,  $\alpha_2$  is the rate coefficient for loss of triplet atoms by ionizing collisions with electrons, and  $\alpha_3$  is the rate coefficient for loss of triplet atoms by mutual triplet-metastable collisions (Ref. 2 and this paper). The rate coefficients  $\alpha_1$  and  $\alpha_2$  are averages over the electronenergy distribution function of the product of cross section and electron speed. For the production of triplet-metastable atoms, a composite cross section<sup>21-23</sup> was used (Fig. 2) which included cross sections for excitation of higher levels of the triplet system which can radiatively decay to the metastable



FIG. 2. Composite excitation function for triplet metastables (Refs. 21-23).

level. Recently published results<sup>24</sup> were used for the triplet ionization cross section. The electronenergy distribution functions used were those of Smit<sup>25</sup>; they have been verified experimentally to a degree and appear to be valid in the ranges of electron densities and values of E/p' encountered here.<sup>26</sup> The calculated values of  $\alpha_1$  and  $\alpha_2$  are given in Figs. 3 and 4. These calculated values of  $\alpha_1$  may be compared to the excitation coefficient  $\epsilon_{met}$  calculated by Corrigan and von Engel.<sup>27</sup> If measured electron drift velocities in helium are used to transform  $\epsilon_{met}$  to the form of  $\alpha_1$  used here, one finds agreement near E/p' = 4.<sup>28</sup> However, our  $\alpha_1$  is about five times greater at E/p' = 10. Some of the discrepancy is due to the different excitation functions used in the calculations.

The loss of triplet-metastable atoms by superelastic collisions with electrons is not included in Eq. (2) since calculations indicate this process should not be important at the values of E/p' encountered here. Certainly the most questionable approximation in Eq. (2) comes from not including interactions between the singlet and triplet systems. The most important such interaction is expected to occur between the singlet- and triplet-metastable



FIG. 4. Calculated ionization-rate coefficient.

state levels. The rate at which singlet-metastable atoms are converted into triplet-metastable atoms by electronic collisions may be calculated by use of the theoretical cross sections given by Morrison and Rudge.<sup>29</sup> Using the fact that the triplet-metastable atoms outnumber the singlet atoms by roughly 3:1 under our experimental conditions (determined by optical-absorption measurements), one finds that this calculated rate is comparable to the triplet-metastable ionization rate  $\alpha_{2n_e}M$ . The *net* effect of the singlet-triplet-metastable conversion should be much less, however, due to the occurrence of conversion in both directions.

The solution of Eq. (2) was performed with the aid of a computer. Radial profiles of triplet metastable density were generated for given values of  $n_e$ , N, and E/p'. The electrons were assumed to be in a fundamental diffusion mode and radial variations of gas density were compensated for by the use of mean values of N and p'.

The results of experimental measurements of triplet-metastable densities are shown in Figs. 5 and 6. Note that under conditions of low excitation.



FIG. 3. Calculated excitation-rate coefficient.



FIG. 5. Measured  $He(2^{3}S)$  density profiles at 3 Torr.



FIG. 6. Measured axial  $\text{He}(2^{3}S)$  density contours. Note that gas pressure is a function of axial electron density when E/p' is held fixed.

when metastable-metastable collisions and ionizing metastable-electron collisions can be neglected, the spatial distribution of the metastable atoms should be directly proportional to that of the electrons. At very high currents, when metastable loss is predominately due to ionizing metastable-metastable collisions, the second and third terms of Eq. (2)should be almost in balance; this should lead to a spatially uniform distribution of metastable atoms. This expected behavior is observed here. It is interesting to note that the triplet-metastable atoms outnumber the electrons by greater than 50:1 under some conditions. This makes triplet loss by mutual triplet collisions an important factor even though the rate coefficient for this process is relatively small. Possible effects due to singlet-triplet-metastable collisions (unevaluated at present) may also be significant, even though the triplets outnumber the singlets by roughly 3:1. The extent of the quantitative agreement of the measurements and calculations is indicated in Fig. 7. The agreement shown is considered to be quite good in light of uncertainties in the cross sections and electronenergy distribution functions used in the calculations. The agreement shows that  $He(2^{3}S)$  densities in active discharges can be predicted with good accuracy from published cross-sectional data and measured electron densities, gas densities, and electric fields.

## IV. ELECTRON TEMPERATURE DECAY

In afterglow helium plasmas, the electron gas gains energy through processes which destroy triplet-metastable atoms. This greatly slows the electron temperature decay, as was suggested by Goldstein<sup>30</sup> and has been observed under various conditons.  $^{4-6, 31}$  In this section we describe the quantitative comparison of measured electron temperature decay and the decay calculated from measured triplet metastable and electron density decays in a 10-Torr afterglow plasma, excited by a 15-mA pulse of  $30-\mu$ sec duration.

The fast electrons produced by ionizing metastable-metastable collisions and superelastic electron-metastable collisions will be slowed down rapidly by collisions with electrons and neutrals. Electrons from the first process will start with about 15 eV of energy. This may be inferred from the work of Hotop and Niehaus<sup>32</sup> in which an energy analysis of electrons resulting from Penning reactions is performed. They find that the ejected electrons possess approximately the maximum energy allowed. Electrons involved in the second process will gain about 20 eV. If one compares the rates at which a fast electron loses energy to the electron  $gas^{33}$  and to the neutrals, one finds that energy loss to the neutrals is dominant above roughly 4 eV for a pressure of 10 Torr and an electron density of 10<sup>11</sup> cm<sup>-3</sup>. Thus, for the range of densities of this experiment, a fast electron resulting from either process will give about

$$\Delta E = 4(\text{eV}) \left( n_e / 10^{11} \,\text{cm}^{-3} \right)^{1/2}$$

of energy to the electron gas.

The electron-energy balance equation for a volume of plasma (neglecting conduction losses) is written as follows:





FIG. 7. Comparison of measured (points) and calculated axial tripletmetastable densities.

where  $\alpha_4$  is the superelastic electron-metastable collision rate coefficient,  $\langle \nu \rangle$  is the collision frequency for momentum transfer, and  $T_g$  is the gas temperature. The energy source term in Eq. (3) indicates that the electron gas gains energy as a result of triplet-metastable-metastable collisions and superelastic collisions of electrons with metastable atoms. The energy-loss term indicates that the electron gas loses energy as a result of elastic electron-neutral or electron-ion collisions; the latter were almost negligible in our plasma.

The rate coefficient  $\alpha_4$  may be calculated from the cross section for the reverse process by the use of microscopic reversibility arguments.<sup>8</sup> Since a variety of results is available in the literature, <sup>6</sup>, <sup>31</sup>, <sup>34</sup> we performed a recalculation of  $\alpha_4$ . Using the work of Schulz and Fox<sup>35</sup> and the crosssection peak of Fleming and Higginson, <sup>22</sup> we find



FIG. 8. Metastable and electron density decays at 10 Torr with the corresponding values of calculated and measured electron temperature.

$$\alpha_4 = (2.7 \times 10^{-11}) T_e^{1/2} \text{ cm}^3 \text{sec}^{-1} (T_e \text{ in } ^\circ\text{K})$$

In order to solve Eq. (3), the time derivative term was dropped since the observed electron temperature decay is much slower than would be expected due to the elastic loss term alone. Spatial distributions appropriate to our plasma (M measured to be parabolic;  $n_e$  assumed to be parabolic) were inserted into the integrals and the integrations were carried out over a section of plasma. With  $T_e$  assumed to spatially uniform, the result in terms of axial metastable and electron densities was

$$(T_e)^{1/2} (T_e - T_g) = (1.4 \times 10^{-24}) \frac{M_0^2}{(n_{e0})^{1/2}} \times \left(1 + \frac{0.03(T_e)^{1/2} n_{eo}}{M_0}\right) , \quad (4)$$

where all units are mks and a pressure of 10 Torr has been used. This equation thus predicts the electron temperature as a function of metastable and electron densities.

The accuracy of Eq. (4) is indicated in Fig. 8; here experimentally measured electron and metastable density decays are shown along with the electron temperatures calculated according to Eq. (4). Good agreement is obtained with the temperatures inferred from collision frequency measurements.

#### V. REMEASUREMENT OF RATE COEFFICIENT FOR IONIZING TRIPLET-METASTABLE-METASTABLE COLLISIONS

The loss of triplet-metastable atoms by mutual metastable collisions which result in the deexcitation of one metastable atom and the ionization of the other is important in both active-discharge and afterglow helium plasmas. The rate coefficient for this process was measured at 300 °K and 10 Torr by Phelps and Molnar<sup>2</sup> and near 520 °K at several pressures by Hurt.<sup>36</sup> Conflicting results were obtained by several workers<sup>37-39</sup> who measured the rate coefficient as a function of pressure in cryogenic plasmas. We report here a measurement of the rate coefficient by a different technique at 300 °K in the pressure range 10-40 Torr.

The afterglow plasmas involved in this part of our work were generated by 1-A 5- $\mu$ sec current pulses. Metastable density measurements were performed during the first 400  $\mu$ sec of the afterglow, when electron temperatures were still high enough so that formation of triplet-metastable atoms by atomic recombination should not have been a significant factor, according to calculations based on the work of Bates, Kingston, and Mc-Whirter.<sup>40</sup> Radial profile measurements of trip-

let densities showed that diffusive loss of metastables at the center of the plasma could be neglected compared to volume losses during the first 400  $\mu$ sec. The radial distribution was observed to be very much flatter than a diffusion distribution, and the diffusive loss from a parabolic distribution would have been always less than a 30%contribution on axis. Loss of triplet atoms in three-body collisions<sup>3</sup> was a significant effect only at the highest pressures and lowest metastable densities encountered here, so that it could be neglected most of the time. Singlet-metastable densities were negligible throughout the measurement times. With these considerations in mind, we write the triplet-metastable density continuity equation for the metastables at the axis of our discharge tube as follows:

$$\frac{dM_0}{dt} = -\alpha_3 M_0^2 - \alpha_4 n_{e0} M_0 = -\alpha_3 M_0^2 \left( 1 + \frac{\alpha_4 n_{e0}}{\alpha_3 M_0} \right) .$$
(5)

In the event that  $\alpha_4 n_{e0} \ll \alpha_3 M_0$ , the equation will have a particularly simple solution; the reciprocal of the metastable density should vary linearly with time, with  $\alpha_3$  as the constant of proportionality. Figure 9 shows the observed behavior at 10 Torr, and Fig. 10 shows the measured rate coefficient taken from data such as shown in Fig. 9 for several pressures. The agreement with Phelps and Molnar's value of  $1.8 \times 10^{-9}$  cm<sup>3</sup> sec<sup>-1</sup> is excellent and the pressure independence is as expected.



FIG. 9. Reciprocal metastable density decay measured at 10 Torr.



FIG. 10. Measured slopes from reciprocal metastable density plots at several pressures.

However, calculations show that for  $T_e$  near 1000 °K, the term  $(\alpha_4 n_{e0} / \alpha_3 M_0)$  is about 20% at all pressures studied here. Furthermore, since this term is not large and is rather slowly varying with time, its presence would not be expected to be visible in a plot such as Fig. 9.

A microwave heating experiment was performed in order to check on the presence of an electron temperature-dependent loss term for triplet-metastable atoms. A microwave signal of sufficient intensity to greatly quench all visible afterglow light was transmitted through the plasma.<sup>30</sup> While the signal was weak enough not to excite detectable numbers of triplet atoms to the singlet-metastable level, it was found that the presence of this signal could enhance the triplet-metastable loss rate by 50% or more at early times in the afterglow, and somewhat less at later times. This is in agreement with the behavior expected from either a superelastic loss term or an atomic recombination source term.

Thus the results of this part of our work suggest a slight downward revision in the triplet-metastable-metastable collision rate coefficient at 300 °K. These results also indicate that further effort might well be spent to clarify this question.

## VI. CONCLUSIONS

In this paper we have reported several measurements of  $He(2^{3}S)$  metastable density behavior which were performed by the use of a laser interferometer in a manner similar to that used previously in neon metastable atom studies.<sup>17, 41, 42</sup> It has been shown that by use of a continuity equation with appropriate source and loss terms and calculated electron-energy distribution functions, the behavior of the triplet-metastable population in active discharges in helium can be described accurately. It also has been shown that, with knowledge of metastable and electron densities, the electron temperature decay in afterglow helium plasmas 9<sup>4</sup>

can be predicted quite accurately. A remeasurement of the rate coefficient for ionizing tripletmetastable-metastable collisions has been reported; good agreement has been obtained with the previous values of Phelps and Molnar, <sup>2</sup> though consideration of effects due to electrons suggests a downward revision of as much as 20%.

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## APPENDIX

Equation (1), which is similar to the results of the standard perturbation theory approach to microwave cavity plasma diagnostics (see, for example, Refs. 43, 44), is derived as follows.

Consider a closed microwave cavity with perfectly conducting walls, but a lossy dielectric filling. It is described by a complex dielectric constant and complex resonant frequency  $\epsilon_1(r)$ and  $\omega_1$ . When a plasma is introduced into the cavity, these quantities change to  $\epsilon_2(r)$  and  $\omega_2$ . The fields in the two cases are denoted by appropriate subscripts. Maxwell's equations for the two cases may be written

$$\nabla \times \overline{\mathbf{E}}_1 = -j\omega_1 \,\mu_0 \,\overline{\mathbf{H}}_1 \,, \tag{A1}$$

$$\nabla \times \vec{\mathbf{H}}_1 = j \,\omega_1 \,\epsilon_1 \, \vec{\mathbf{E}}_1 \,, \tag{A2}$$

 $\nabla \times \vec{\mathbf{E}}_2 = -j\omega_2 \,\mu_0 \vec{\mathbf{H}}_2 \,, \tag{A3}$ 

$$\nabla \times \vec{\mathbf{H}}_2 = j\omega_2 \epsilon_2 \vec{\mathbf{E}}_2 . \tag{A4}$$

If we conjugate the first two equations, multiply the equations by  $\vec{H}_2$ ,  $\vec{E}_2$ ,  $\vec{H}_1^*$ , and  $\vec{E}_1^*$ , respectively, subtract the equations from each other in the appropriate fashion, and perform the customary volume integration, we find that

$$\frac{\omega_a - \omega_b}{\omega_b} = \frac{1}{2} j \frac{\omega_a}{\omega_b} \left( \frac{1}{Q_2} + \frac{1}{Q_1} \right)$$
$$= \left( \int \Delta \epsilon_p \vec{\mathbf{E}}_1^* \cdot \vec{\mathbf{E}}_2 d^3 r + 2j \int \epsilon_{1i} \vec{\mathbf{E}}_1^* \cdot \vec{\mathbf{E}}_2 d^3 r \right) \Big/$$

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$$\int \left(\epsilon_1^* \vec{\mathbf{E}}_1^* \cdot \vec{\mathbf{E}}_2 + \mu_0 \vec{\mathbf{H}}_1^* \cdot \vec{\mathbf{H}}_2\right) d^3 r , \qquad (A5)$$

where  $\omega_a$  and  $\omega_b$  are the real parts of  $\omega_1$  and  $\omega_2$ ,  $\Delta \epsilon_p(r)$  is the complex change in dielectric constant caused by the plasma, and  $\epsilon_{1i}(r)$  is the imaginary part of  $\epsilon_1(r)$ .

As the fields  $\vec{E}_2$  and  $\vec{H}_2$  approach  $\vec{E}_1$  and  $\vec{H}_1$ , the latter ratio of integrals approaches  $(-2Q_1)^{-1}$ . If either  $Q_1$  is considerably larger than  $Q_2$  or  $\vec{E}_1$  is approximately equal to  $\vec{E}_2$ , then the error introduced by replacing this ratio by  $(-2Q_1)^{-1}$  will be very small. Using this substitution, we get

$$\frac{\Delta\omega}{\omega_b} - \frac{1}{2}j \frac{\omega_a}{\omega_b} \Delta \left(\frac{1}{Q}\right) = \int \Delta \epsilon_p \vec{\mathbf{E}}_1^* \cdot \vec{\mathbf{E}}_2 d^3 r / \int (\epsilon_1^* \vec{\mathbf{E}}_1^* \cdot \vec{\mathbf{E}}_2 + \mu_0 \vec{\mathbf{H}}_1^* \cdot \vec{\mathbf{H}}_2) d^3 r , \quad (A6)$$

where  $\Delta \omega = \omega_a - \omega_b$  and

$$\Delta\left(\frac{1}{Q}\right) = \frac{1}{Q_2} - \frac{1}{Q_1}\left(\frac{2\omega_b}{\omega_a} - 1\right) \approx \frac{1}{Q_2} - \frac{1}{Q_1} \quad . \quad (A7)$$

This last approximation would be good if either  $\omega_a = \omega_b$  or  $Q_2 \ll Q_1$ .

If electron-neutral collisions predominate, then  $\Delta \epsilon_{p}$  is spatially uniform and the ratio of the imaginary part to the real part of  $\Delta \epsilon_{p}$  may be taken as  $\langle \nu \rangle / \omega_{b}$ , to a good approximation.<sup>12</sup> The ratio of integrals in Eq. (A6) may be separated into real and imaginary parts easily, assuming that the denominator is approximately real (questionable at very high collision frequencies). By taking the ratios of real to imaginary parts of both sides of Eq. (A6) and equating them, one finds that

$$\langle \nu \rangle = \omega_a \omega_b \Delta (1/Q)/2 \left| \Delta \omega \right| . \tag{A8}$$

Thus the collision frequency can be determined approximately from measurements of the shift in resonant frequency of a well-undercoupled cavity and the change in cavity Q caused by the plasma, without evaluating any integrals or knowing any of the fields.

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#### PHYSICAL REVIEW A

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# Scattering Theory and Current Correlations in Classical Gases

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We study the space-time-dependent correlation functions of density and velocity in a dilute gas using classical scattering theory. A density expansion of correlation functions is discussed. The effect of two-body interaction is analyzed in detail for the case where the relevant wavelength is long compared to the atomic dimension but small compared to the mean free path. Explicit formulas for the correlation functions in terms of the impact parameter and the time delay are obtained. The equivalent quantum-mechanical analysis has been carried out elsewhere. The classical and quantum formulas are nearly identical. A main purpose of this paper is to make the results of the (difficult) quantum-mechanical treatment more accessible.

## I. INTRODUCTION

In a recent paper, <sup>1</sup> we investigated the correlation functions of gaseous systems from the viewpoint of quantum scattering theory. The present paper is devoted to the same topic using classical rather than quantum mechanics. The classical treatment is vastly simpler, both conceptually and in technical detail. In fact, a major purpose of this paper is to make the ideas of Ref. 1 accessible to a larger class of readers. Also, in many cases where one might want to apply the results of Ref. 1, classical mechanics is entirely adequate.

The main point of our quantum-mechanical investigation in Ref. 1 was that in an interesting nontrivial regime of low frequencies and long wavelengths, the current correlation functions of a gas can be expressed in terms of quantities directly measurable in scattering experiments. By a scattering experiment we mean, of course, one where only the asymptotic (straight-line) trajectories of particles before and after collisions are observed.

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