Effect of the Imprisonment of Resonance Radiation on Excitation Experiments

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The theory of the imprisonment of resonance radiation developed by Holstein and by Bieberman is applied to the analysis of experimental measurements of excitation cross sections for the resonance ($n\,^1P$) states of helium. The transport equation for the density of atoms in the resonance state is solved numerically for the case of a thin sheet of exciting electrons between parallel plane electrodes. The fraction of the resonance atoms producing visible radiation is then calculated for parallel plane geometry and estimated to within five percent for cylindrical geometry with an axial electron beam. Predictions of the theory are compared with the available experimental data for helium. The theory shows that at the helium pressures commonly used (10^{-3} to 10^{-1} mm of Hg) the observed visible radiation may easily be a factor of ten greater than that expected when imprisonment effects are neglected. As a result, the cross sections for the excitation of the $n\,^1P$ states given in the literature are much too large. For example, our analysis of the available experimental data suggests that the cross section for excitation by electrons to the $3\,^1P$ state of helium at 100 electron volts is 3×10^{-18} cm^2 instead of 4×10^{-17} cm^2 as given in the literature.

I. INTRODUCTION

THE object of this paper is to apply the theory of the imprisonment of resonance radiation developed by Holstein[1,2] and by Bieberman[3] to the analysis of experimental measurements of the excitation cross sections for resonance states of atoms. We will be concerned with those resonance states which can radiate to one or more excited states as well as to the ground state of the atom. Because of the large number of atoms in the ground state in an excitation experiment, the radiation emitted in a transition to the ground state will be absorbed and re-emitted many times before reaching the wall of the tube. The resultant increase in the effective lifetime of the excited state against radiation to the ground state, known as the "imprisonment of resonance radiation," results in an increase in the probability that an excited atom will decay to a lower "nonresonant" excited state with the emission of visible or infrared radiation. The $(1s\,np)^1P$ states of helium are examples of this class of excited states and have been studied by a number of experimenters.[4–7] In general, the experimental arrangement consists of an electron beam passing along the axis of a short cylindrical collision chamber. We will assume that the optical system is designed to allow a determination of the total radiation emitted in a thin slab perpendicular to the axis of the electron beam.[4] Although the imprisonment of resonance radiation was shown by Lees and Skinner[8] to result in a spreading of the spatial distribution of excited atoms in the collision chamber and a variation in the observed excitation cross section with pressure, Heddle[9] was the first to show how one could take into account empirically the effects of imprisonment on the measured cross sections.

In the second section of this paper we will derive the relationship between the experimentally measured intensity of radiation emitted in transitions between excited states and any given distribution of the density of excited atoms. The Holstein-Bieberman theory will then be used to find an equation for the spatial distribution of the excited atoms. In the third section we will solve for the excited atom density and the fraction of emitted "visible" radiation for the case of parallel plane geometry. In the fourth section we will obtain maximum and minimum values for the fraction of radiation appearing as visible radiation for infinite parallel plane and cylindrical collision chambers. These limiting values can be used to analyze experiments for which the exact density distribution has not or cannot be found. In the last section some of the predictions of the theory are compared with published experimental data.

II. THEORY

The measured quantities in an excitation experiment are the intensity of radiation in a given spectral line

[1] T. Holstein, Phys. Rev. **72**, 1212 (1947). Experimental confirmation of the theory of the decay of resonance radiation is given by Alpert, McCoubrey, and Holstein, Phys. Rev. **76**, 1257 (1949); Holstein, Alpert, and McCoubrey, Phys. Rev. **85**, 985 (1952); and A. V. Phelps and A. O. McCoubrey, Bull. Am. Phys. Soc. Ser. II, **3**, 83 (1958).

[2] T. Holstein, Phys. Rev. **83**, 1159 (1951). The values of $g_L(k_0L)$ and $g_R(k_0R)$ given in Fig. 4 are some 30% larger than those given in this reference. The difference results from the use of a more accurate $K(k_0\rho)$ function. Note that the coefficient A in Eq. (5) could include any volume destruction process for the resonance states, e.g., collisional de-excitation.

[3] L. M. Bieberman, J. Exptl. Theoret. Phys. U.S.S.R. **17**, 416 (1947).

[4] J. H. Lees, Proc. Roy. Soc. (London) **A137**, 173 (1932).

[5] O. Thieme, Z. Physik **78**, 412 (1932); W. Hanle and W. Schaffernicht, Ann. Physik **6**, 905 (1930).

[6] H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Oxford University Press, London, 1952), Chaps. II and III.

[7] H. S. W. Massey, *Handbuch der Physik* (Springer-Verlag, Berlin, 1956), Vol. 36, p. 325; and R. G. Fowler, *Handbuch der Physik* (Springer-Verlag, Berlin, 1956), Vol. 22, p. 209.

[8] J. H. Lees and H. W. B. Skinner, Proc. Roy. Soc. (London) **A137**, 186 (1932).

[9] D. W. O. Heddle, Warren Conference Report on Physics of Ionized Gases, University of Birmingham, July, 1954 (unpublished), p. 41. The author states (private communication) that the experimental data should be regarded as preliminary and that the final cross sections for the 5016A line are roughly twice the values given.

emitted from the tube, the electron current through the tube, the gas density, and the dimensions of the electron beam and the collision chamber. The rate of visible photon emission per unit length of electron beam, J_{jk}, is

$$J_{jk}=A_{jk}\int n(\mathbf{r})da, \qquad (1)$$

where A_{jk} is the transition probability per unit time for the experimentally observed line, $n(\mathbf{r})$ is the density of excited atoms at the point $\mathbf{r}$, and da is an element of area perpendicular to the electron beam. The rate of production of excited atoms per unit length by direct excitation[10] is

$$R_j=\int p(\mathbf{r})da\equiv NQ_jI/e, \qquad (2)$$

where N is the gas density, Q_j is the excitation cross section for the jth state, $p(\mathbf{r})$ is the rate of production of excited atoms per unit volume, I is the electron current, and e is the electronic charge. In this discussion, we are concerned with the fraction of the excited atoms which result in visible photons, i.e.,

$$f_{jk}=J_{jk}/R_j=A_{jk}\int n(\mathbf{r})da\bigg/\int p(\mathbf{r})da. \qquad (3)$$

The quantity f_{jk} will vary from a small number of the order of 10^{-2} at low gas densities to a value as large as unity at high gas densities. It is often convenient to discuss the experimental results in terms of the apparent cross section, Q_{jk}, for the production of a particular line. The apparent cross section is defined by the equation

$$Q_{jk}=eJ_{jk}/NI,$$

and is therefore related to the total cross section[10] by

$$Q_{jk}=f_{jk}Q_j. \qquad (4)$$

If we designate the sum of the Einstein coefficients for all lines other than the resonance line by A and that for the resonance line by γ, the density of excited states, $n(\mathbf{r})$, satisfies the Boltzmann transport equation[1,2]

$$p(\mathbf{r})+\gamma\int n(\mathbf{r}')G(|\mathbf{r}-\mathbf{r}'|)dv'-\gamma n(\mathbf{r})-An(\mathbf{r})=0, \qquad (5)$$

where $p(\mathbf{r})$ is the rate of production of excited atoms per unit volume and $G(|\mathbf{r}-\mathbf{r}'|)$ is the probability that the radiation emitted by an atom at $\mathbf{r}'$ will be absorbed by an atom at $\mathbf{r}$. The second term gives the rate of absorption at $\mathbf{r}$ while the last two terms give the rate of emission at $\mathbf{r}$. Over much of the range of gas densities and tube sizes of interest, the second and third terms nearly cancel. Because of the slow decrease in $G(|\mathbf{r}-\mathbf{r}'|)$ with $|\mathbf{r}-\mathbf{r}'|$ for this problem, Eq. (5) cannot be reduced to a diffusion equation, nor can one define a mean free path for the photons.[1,3]

The dependence of the quantity $G(|\mathbf{r}-\mathbf{r}'|)$ upon the distance $|\mathbf{r}-\mathbf{r}'|$ and the density of absorbing atoms depends upon the details of the spectral line. In general,[1] $G(\rho)=(1/4\pi\rho^2)dT(\rho)/d\rho$, where $T(\rho)=\int_{-\infty}^{\infty}\epsilon(\nu)e^{-\rho k(\nu)}d\nu$ and $\rho=|\mathbf{r}-\mathbf{r}'|$. Here $T(\rho)$ is the probability that the radiation emitted by an atom will travel a distance ρ before absorption, $\epsilon(\nu)$ and $k(\nu)$ are the frequency-dependent coefficients of emission and absorption, and ν is the frequency of the radiation. Holstein has evaluated $T(\rho)$ for large ρ in the cases of Doppler,[1] impact, and statistical broadening.[2] He assumes, as does Bieberman, that the spectral distribution of emission and absorption are identical and discusses the significance of the assumption in detail. Bieberman[3] has treated the Doppler case for all values of ρ. As discussed by Holstein,[1] the analysis presented here is not valid when natural broadening is important.

The case of combined Doppler and impact broadening for large ρ has been considered by Walsh[11] and by Bieberman and Gourevitch.[12] It is assumed that in the range of interest the spectral distribution of the absorption coefficient is given by[13]

$$k(\omega)=k_0[\exp(-\omega^2)+a/\pi^{\frac{1}{2}}\omega^2], \qquad (6)$$

where

$$k_0=\frac{\lambda^3N}{8\pi^{\frac{3}{2}}}\frac{g_2}{g_1}\frac{\gamma}{v_0},$$

$\omega=\Delta\nu\lambda/v_0$, $v_0=(2kT/M)^{\frac{1}{2}}$, and $a=(\gamma+\gamma_p)\lambda/4\pi v_0$. Here k_0 is the absorption coefficient of the center of the Doppler-broadened line, λ is the wavelength, N is the density of atoms in the ground state, g_2 and g_1 are the statistical weights of the upper and lower states, $\Delta\nu$ is the frequency as measured from the center of the line, k is Boltzmann's constant, T is the gas temperature, M is the mass of the gas atoms, and γ_p is a pressure-broadening coefficient approximately equal to the frequency of collisions between excited atoms and normal atoms.[3] Equation (6) is valid only for $a\ll1$. Because of natural broadening, the derived values of $T(\rho)$ are useful only for $\gamma\ll\nu_c+4\pi^{\frac{3}{2}}\lambda^{-1}v_0\omega_m^2\exp(-\omega_m^{-2})$, where ω_m is the frequency at which $\epsilon(\omega)e^{-\rho k(\omega)}$ is a maximum. At large values of $k_0\rho$ the results for $T(\rho)$ are the same as those obtained by Holstein for the impact-broadening case. At small values of $k_0\rho$ and $a=0$, we use the

[10] Here we have neglected the effects of cascading from higher states and the transfer of excitation. When these effects are not small, one must solve an involved set of coupled differential equations. Because of these effects, the theoretical and experimental spatial distributions can be compared only under ideal conditions. See Sec. V for a calculation of the effects of the transfer of excitation on the apparent cross section, Q_{jk}. Fortunately, the effects of cascading are generally small and can be taken into account without solving the coupled equations (reference 6).

[11] P. J. Walsh (private communication).

[12] L. M. Bieberman and I. M. Gourevitch, J. Exptl. Theoret. Phys. U.S.S.R. **20**, 108 (1950).

[13] A. G. C. Mitchell and M. W. Zemansky, *Resonance Radiation and Excited Atoms* (The Macmillan Company, New York, 1934), p. 323.

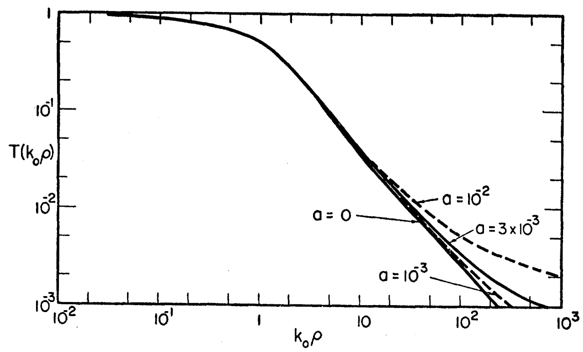

FIG. 1. Transmission function for Doppler- and impact-broadened radiation. This function gives the probability that a photon will travel a distance of $k_0\rho$ without absorption. It is assumed that spectral distribution of the emission probability and the absorption coefficient are the same.

relation $T(k_0\rho)=1-A_1(k_0\rho)$ where A_1 is the absorption factor, A_α, defined by Mitchell and Zemansky,[13] for $\alpha=1$. Figure 1 shows the values of $T(k_0\rho)$ obtained for various values of a using Walsh's formula. Using the relation $a=\lambda\gamma/4\pi v_0+\lambda k_0/4\pi^{\frac{3}{2}}$ for the resonance state,[1] we find that for $k_0<10^3$ and the $n\,^1P$ states of helium $(n\geq3)$, $a<3\times10^{-3}$. Since $\rho\leq1$ cm and accurate values of $T(k_0\rho)$ will be required only for $k_0\rho<10^2$, pressure-broadening effects will be neglected in our calculations of f_{jk}. At high gas densities, pressure-broadening effects make significant contributions to the quantity $(1-f_{jk})$ for all states and to f_{jk} for the $2\,^1P$ state. At helium densities above about 10^{15} atom/cc and below about 10^{18} atom/cc, our present analysis cannot be applied to the $2\,^1P$ state because of large natural-broadening effects.

Equation (5) has been solved numerically in two cases. Bieberman considered the situation in which the production term, $p(\mathbf{r})$, was due to a beam of resonance radiation incident on an absorbing gas contained between infinite parallel planes. He calculated the resultant spatial distributions of the excited-atom density for values of $k_0L<10$, where L is the separation between plates. Siewert[14] has examined the same problem for large k_0L and a somewhat more complicated geometry. An equation similar to Eq. (5) has been obtained by Holstein,[1] and solved by Walsh[15] from a consideration of the escape of resonance radiation when there is an appreciable probability of de-excitation to the ground state of the atom by collisions with electrons, in addition to the production of resonance atoms by electron excitation. Walsh calculated the two lowest eigenfunctions and then applied perturbation theory to find the resultant spatial distribution and rate of loss of resonance atoms.

III. PARALLEL PLANE EXCITATION TUBE

In this section we will solve for the spatial distribution of excited atoms and the values of f_{jk} which result when the electron beam is confined to a sheet of thickness δ located midway between infinite parallel plane electrodes separated by a distance L. In this case the density is a function only of a single coordinate z. Fortunately, the other coordinates are easy to eliminate. Thus, Bieberman shows that for a Doppler-broadened line,

$$K(|z-z'|)=\int G(\rho)dx'dy' =\frac{k_0}{2(\pi)^{\frac{1}{2}}}\int_{-\infty}^{\infty}\exp(-2\omega^2)d\omega\int_{k_0\rho\exp(-\omega^2)}^{\infty}\frac{e^{-t}dt}{t}, \quad (7)$$

where $\rho=|z-z'|$. Holstein shows that for large ρ

$$K(|z-z'|)=\int G(\rho)dx'dy' =\frac{ma_m}{2(m+1)\rho^{m+1}}=\frac{mT(\rho)}{2(m+1)\rho}, \quad (8)$$

where $T(\rho)$ is approximated by $T(\rho)=a_m/\rho^m$. The values obtained by applying Holstein's approximate formula agree well with those tabulated by Bieberman for values of $k_0\rho>1$, provided that one allows values of m greater than unity.

Our problem now is to solve the equation

$$p(z)+\gamma\int_{-\frac{1}{2}k_0L}^{\frac{1}{2}k_0L}n(z')K(|z-z'|)dz'-\gamma n(z)-An(z)=0. \quad (9)$$

In this case,

$$f_{jk}=A_{jk}\int n(z)dz\bigg/\int p(z)dz. \quad (10)$$

Following Bieberman's second method of solution,[3] we

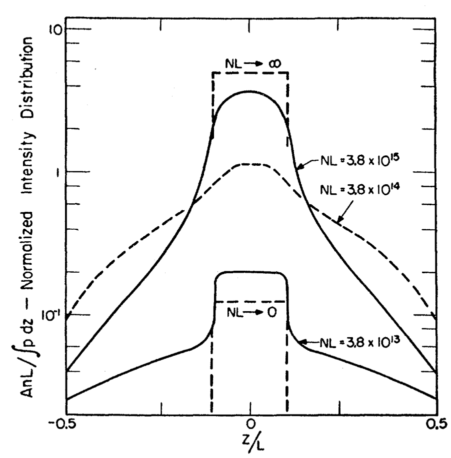

FIG. 2. Theoretical normalized excited atom density and visible intensity distribution for the helium $3\,^1P$ state and parallel-plane geometry. These results will also apply to any excited state for which $A/\gamma=0.024$, provided that the parameter NL is replaced by the parameter k_0L. Starting with the upper curve, the values are $k_0L\rightarrow\infty$, $k_0L=200$, 20, and 2, and $k_0L\rightarrow0$, respectively.

[14] R. Siewert, Ann. Physik **17**, 371 (1956).
[15] P. J. Walsh, Phys. Rev. **107**, 338 (1957).

can write the integral equation as a system of algebraic equations as follows:

$$P_i = n_i\Gamma - \sum_{l=1}^{q} \theta_{i-l} n_i, \tag{11}$$

where $\Gamma = (\gamma + A)/\gamma$, $P_i = p(z_i)/\gamma$,

$$\theta_m = \int_{(2m-1)L/2q}^{(2m+1)L/2q} K(|z|)dz,$$

and $m = |i-l|$. The above solution gives values of n at q points separated by a distance of L/q. The end points are located at a distance of $L/2q$ from the absorbing walls at $z = \pm L/2$.

Figure 2 shows plots of $An(z)L/\int p(z)dz$ as calculated for the $3\,^1P$ state of helium[16] for $\delta/L = 0.2$ and various values of NL ($q = 20$). These curves are plotted such that the ordinate is proportional to the visible radiation intensity which would be observed to originate from a thin slab at z, if the gas density were changed and the total rate of production of excited atoms kept constant by varying the electron current. For the limiting values of $NL = 0$ and $NL = \infty$, the spatial distribution of excited atoms is the same as the spatial distribution of the electrons, i.e., constant between $z = \pm\delta/2$. At the low-density limit the resonance radiation reaches the wall of the tube without absorption. At the high-density limit the resonance radiation is absorbed so strongly that the atoms radiate to a lower excited state before the resonance radiation can reach a point appreciably outside the production region. At intermediate gas densities the density of resonance atoms outside the production region is appreciable as observed by Lees and Skinner.[8] Calculations of $n(z)$ were also made for $\delta/L = 0.1$ and 0.3. When normalized to the same total production, the density near the walls was found to be very nearly the same as in Fig. 2, while the extent of the high-density region near the center changed with δ/L in such a way as to keep the total area under the curve essentially constant. The solid curve of Fig. 3 shows the values of f_{5016} *vs* k_0L obtained by integration of the curves of $n(z)$. The values of f_{5016} computed for $\delta/L = 0.1$, 0.2, and 0.3 were equal to within 1%.

IV. MAXIMUM AND MINIMUM VALUES OF f_{jk}

In this section we shall obtain limiting values for the fraction of the excited atoms producing visible photons.

[16] The values of A_{5016} and γ used are those used by Heron, McWhirter, and Rhoderick, Proc. Roy. Soc. (London) **A234**, 565 (1956). The values of γg obtained by subtracting $A_{5016} = 1.35\times10^7$ sec^{-1} from the values of $1/\tau$ measured in this paper show the same variation with k_0R as shown in our Fig. 5, but seem to require an effective radius 40% larger than the actual radius. Reflection of the resonance radiation at the quartz wall is not a satisfactory explanation for this discrepancy since the measured reflection coefficient for 600A radiation is 7.5% and would lead to approximately the same percentage change in γg. See P. R. Gleason, Proc. Natl. Acad. Sci. U. S. **15**, 551 (1929).

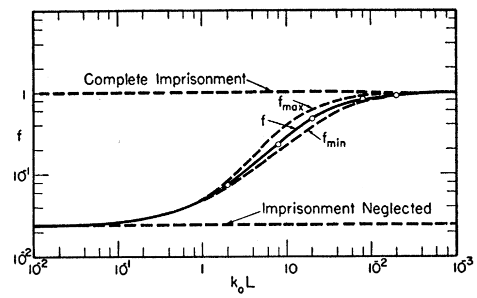

FIG. 3. Theoretical curves of the fraction of the excited helium $3\,^1P$ atoms which emit visible photons in parallel-plane geometry. The points indicated by circles (○) are those obtained by integration of curves such as shown in Fig. 2. These results apply to any state for which $A/\gamma = 0.024$. For the helium $3\,^1P$ level, $NL = 1.9\times10^{13} k_0 L$ at 300°K.

Since these limits can be computed for both cylindrical and parallel plane geometries, we can make use of a comparison of the limits with the exact value in the parallel plane case in order to arrive at a reasonably accurate value of f_{jk} for cylindrical geometry. Such a procedure is the best we can do, since we have been unable to reduce Eq. (5) to the form of Eq. (9) in the cylindrical case. In addition, these limits can be used to calculate values of f_{jk} for analyzing data where the parameters, or even the differential equation, differ from those considered so far.

An upper limit of f_{jk} is obtained by first integrating Eq. (5) over the volume of the collision chamber. This procedure takes advantage of the fact that most of the emitted resonance radiation is reabsorbed within the collision chamber and so drops out of the integrated equation leaving only the radiation absorbed external to the gas in the collision chamber. Solving for f_{jk}, we obtain the expression

$$f_{jk} = A_{jk} \Big/ \left[A + \frac{\gamma \int n(\mathbf{r}')dv' \int_{\text{ext}} G(|\mathbf{r}-\mathbf{r}'|)dv}{\int n(\mathbf{r})dv} \right]. \tag{12}$$

Here we have made use of the fact that the integral of $G(|\mathbf{r}-\mathbf{r}'|)$ over all space is unity, i.e.,

$$1 - \int_{\text{int}} G(|\mathbf{r}-\mathbf{r}'|)dv = \int_{\text{ext}} G(|\mathbf{r}-\mathbf{r}'|)dv, \tag{13}$$

where the integrals are evaluated over the volumes internal to and external to the collision chamber. An upper limit of f_{jk} is obtained when the second term in the denominator of Eq. (12) is made smaller than the true value. Since $G(|\mathbf{r}-\mathbf{r}'|)$ decreases with increasing $|\mathbf{r}-\mathbf{r}'|$ this is accomplished by replacing $G(|\mathbf{r}-\mathbf{r}'|)$ by $G(|\mathbf{r}|)$, i.e., the value of G which is appropriate for excited atoms located at the center of the collision chamber. With this assumption, we obtain

$$f_{jk\,\max} = A_{jk}/(A + \gamma s), \tag{14}$$

where $s = \int_{\text{ext}} G(|\mathbf{r}|)dv$ and is to be evaluated at all points outside the collision chamber. Figure 4 shows

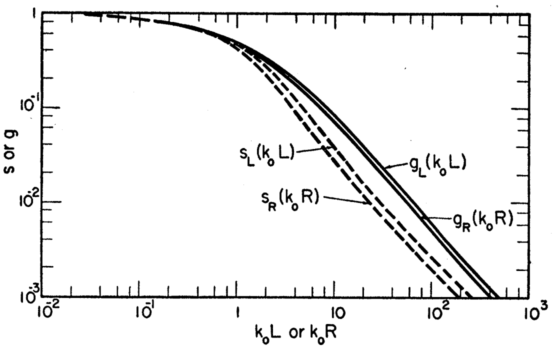

FIG. 4. Imprisonment coefficients for parallel-plane and cylindrical geometries for a Doppler-broadened line. The subscript L refers to the parallel-plane geometry and the subscript R refers to cylindrical geometry. The coefficient s is the probability that radiation emitted from the center of the tube will reach the wall without absorption. The product of the coefficient g and the resonance transition probability γ is the decay constant for resonance radiation distribution spatially in the fundamental decay mode, so long as $\gamma g \gg A$. For smaller γg, the decay constant is $\gamma g + A$. A comparison of Figs. 1 and 4 shows that the s and g coefficients are within a factor of two of $T(k_0\rho)$, where ρ is R or $L/2$ for cylindrical and parallel-plane geometries, respectively.

the values of s calculated for parallel plane and cylindrical geometry for the 3 ^1P state of helium. Figure 3 shows a comparison of $f_{jk\,\max}$ with the "exact" value.

A lower limit to f_{jk} is obtained by assuming that the spatial distribution of the production term and, therefore, the excited atoms, is the same as that of the fundamental decay mode obtained by Holstein. Walsh[15] shows how the spatial distribution of the fundamental mode varies with k_0L. In this limit, Eq. (5) becomes

$$p(\mathbf{r}) = \gamma g n(\mathbf{r}) + A n(\mathbf{r}),$$

so that

$$f_{jk\,\min} = A_{jk}(A+\gamma g)^{-1}, \tag{15}$$

where γg is the decay constant characteristic of the fundamental decay mode. It should be noted that Eq. (15) does not give a true minimum for f_{jk}, since a production term which is relatively larger near the walls will lead to smaller f_{jk} values. However, this lower limit should be satisfactory for most experiments, since the electron current is concentrated near the center of the tube. Holstein shows how to determine g from $T(k_0\rho)$ by using a variational technique.[1,2] Figure 4 shows the results of an application of this technique to the calculation of $g_L(k_0L)$ for plane parallel geometry using Bieberman's $G(k_0\rho)$ function. Figure 3 shows a comparison of $f_{jk\,\min}$ with the exact value. Figure 4 also shows values of $g_R(k_0R)$ for large k_0R obtained by using Eq. (5.33) of reference 2. At $k_0R<2$ our curve of $g_R(k_0R)$ is an estimate based on the $g_L(k_0L)$ curve and is believed to be correct to better than 10%. Rather than calculate $f_{\max}$ and $f_{\min}$, we can calculate an effective f_{jk} using the average value of s and g instead of s in Eq. (14). In the case of parallel plane geometry, Fig. 3, the value of f_{5016} calculated in this way agrees with the exact value to within about 5%.

V. APPLICATION OF THEORY TO EXPERIMENT

A direct comparison of our theory with experimental data is difficult since most of the measurements of apparent cross sections have been carried out in geometries which are neither parallel-plane nor cylindrical. Because of this, there is little point in comparing our calculated distribution of excited atoms with that observed by Lees and Skinner.[8] Fortunately, our theory shows that the f_{jk} curves have almost exactly the same shape for parallel-plane and cylindrical geometries. This suggests that we analyze the experimentally determined cross-section data for any geometry using either theoretical curve and Eq. (4), and then comment as to whether the inferred effective dimension of the collision chamber is reasonable. Figure 5 shows such a fit of Eq. (4) to the experimental data obtained by Lees[4] and by Thieme[5] for the cross sections for the production of 5016 A radiation at various gas densities. The points shown for Lees are the measured values multiplied by 3. This factor is necessary to bring into agreement the two sets of peak values of excitation cross sections for the $n\,^1S$ and $n\,^3S$ states. These states were chosen for comparison since they are least apt to be pressure-dependent as a result of the collisional transfer of excitation from the $n\,^1P$ and $n\,^1D$ states.[8,17] The correction factor was applied to Lees' data, rather than Thieme's, since this resulted in good agreement with the unpublished measurements of Heddle.[9] The adjustable parameters in the theory are the effective collision-chamber radius and the total cross section for the direct production of atoms in the 3 ^1P state, i.e., Q_j of Eq. (4). The effective radius used to calculate the theoretical curve of Fig. 5 was 0.75 cm. The collision

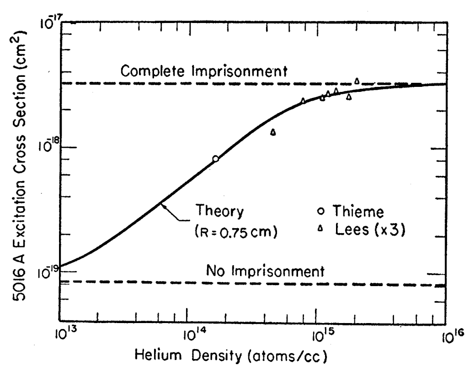

FIG. 5. Fit of theory to measured cross section for the production of the 5016A line of helium by 100-ev electrons. Thieme's experimental point is shown by a circle (○). As discussed in the text, Lees' points are multiplied by three and shown by triangles (△). Equation (4) was fitted to the experimental data by using the average of g_R and s_R from Fig. 4 to calculate $f_{jk}(k_0R)$, an effective radius of 0.75 cm, and a cross section of 3.3×10^{-18} cm² for the direct excitation of the 3 ^1P state.

[17] R. Wolf and W. Mauer, Z. Physik **115**, 410 (1940). See also reference 6, pp. 428–31. The data of reference 16 support our belief that the cross section for excitation transfer from the 3 ^1P state to the 3 ^1D and 3 ^3D states is small, since the observed decay of 3 ^1P density is accounted for by radiation alone.

chamber in Lees' tube was about 4 cm in diameter and 0.8 cm in length. If Thieme's collision chamber is the same as that used by Hanle and Schaffernicht,[5] as is commonly assumed,[6] the dimensions are about 10% larger than those in Lees' tube and the difference can be neglected in this analysis. In view of the small ratio of length to diameter in these collision chambers as compared to the infinite cylinder assumed in our theory, we can make only the qualitative remark that an intermediate value for the effective diameter is reasonable. The excitation cross section obtained by the approximate analysis of Fig. 5, i.e., 3.3×10^{-18} cm^2, is considerably smaller than the value given in the literature,[6] 4×10^{-17} cm^2, and is in good agreement with the prediction of theory,[18] i.e., 3.8×10^{-18} cm^2. In the above analysis we have neglected the effects of the transfer of excitation. We believe that this effect is small for the $3\,^1P$ level, although the effect on the apparent cross section for the high $n\,^1P$ levels is very large.[8,9,17]

As our second example, we will consider the apparent increase with helium density in the cross section for the production of metastables observed by Dorrestein.[19] The circles in Fig. 6 show the experimentally determined metastable excitation cross section[20] at 100 electron volts. The lower solid line shows the sum of the Q_{jk} values for transitions ending on the $2\,^1S$ metastable state as calculated using Eq. (4) under the assumptions that (a) the cross sections for direct excitation to the $n\,^1P$ states at 100 electron volts, $Q(n\,^1P)$, vary inversely as the third power of the principal quantum number,[21] (b) production of metastables between the collision chamber and the detector is negligible, and (c) the effective radius of the collision chamber is equal to the actual radius, i.e., 1 cm. In order to make the slope of the lower solid line equal to that of the line drawn through the experimental points, the total cross section for the excitation of all singlet states except the $2\,^1S$ state was assumed to be 1.3×10^{-17} cm^2, with 10% of

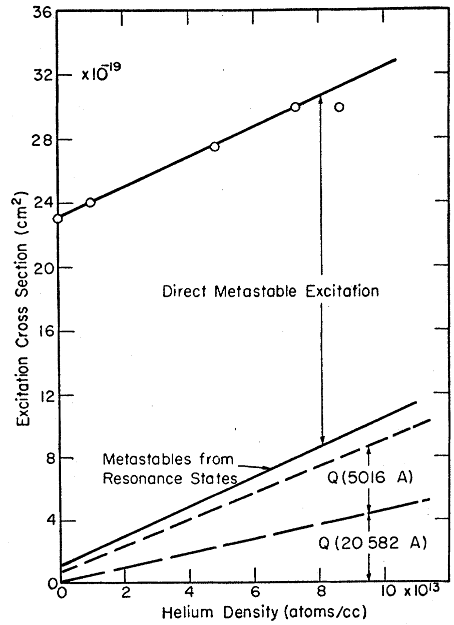

FIG. 6. Separation of the helium metastable production observed by Dorrestein (○) into the metastables produced by direct excitation and those produced by decay from the resonance states.

this cascading directly to the $2\,^1P$ state.[6] The approximate equality of the $2\,^1P$ and $3\,^1P$ contributions as shown by the dashed lines, in spite of the much larger $2\,^1P$ cross section, is the result of the relatively low transition probability for the $2\,^1P-2\,^1S$ transition.[22] The upper straight line through the experimental points is obtained by adding to the $\sum Q_{jk}$ line a constant cross section of 2.2×10^{-18} cm^2 representing the direct production of singlet metastables and the total production of atoms in the triplet states. The analysis of Dorrestein's data leads to a total cross section for the excitation of the $n\,^1P$ states of about 1.2×10^{-17} cm^2 as compared to the theoretical value[21] of 1.5×10^{-17} cm^2. The derived cross sections for the direct excitation of the $3\,^1P$ and $5\,^1P$ states are 2.2×10^{-18} cm^2 and 5×10^{-19} cm^2, respectively. The $3\,^1P$ value is in satisfactory agreement with the value of 3.3×10^{-18} cm^2 suggested by the analysis of Fig. 5. Unfortunately, the nearly linear relation between metastable production and the product of cross section and k_0R at low densities means that one could have used a smaller effective radius and obtained a correspondingly larger excitation cross section for the $3\,^1P$ state.[9] Thus, Dorrestein's experiment does not allow us to decide as to the correct effective radius. However, we feel that the geometry is sufficiently close to cylindrical so that the effective radius should be very nearly equal to the actual radius and that the excitation cross sections are approximately as given. It is important to note that although this type of experiment does not allow us to separate the

[18] See reference 6, Fig. 80(a) and Sec. 3.521. D. R. Bates *et al.*, Trans. Roy. Soc. (London) **A243**, 93 (1950).

[19] R. Dorrestein, Physica **9**, 447 (1942), Fig. 1. Following L. J. Varnerin, Jr., Phys. Rev. **91**, 859 (1953), and H. D. Hagstrum, Phys. Rev. **96**, 336 (1954), we have assumed that both metastable states of helium have the same efficiency for the production of electrons at the platinum surface. On the basis of Meir-Leibnitz' measurements [Z. Physik **95**, 499 (1935)], we have taken the efficiency of electron production by metastables to be 0.20, whereas Dorrestein used 0.24 for the triplet metastable and 0.40 for the singlet metastable.

[20] Dorrestein's apparent metastable cross sections at 100 electron volts were corrected for loss of metastables by scattering by assuming that the decrease with pressure in the apparent cross section for 27-ev electrons is due to scattering. The production of metastables from the resonance states should be negligible at this low electron energy. The low apparent cross section for metastable scattering is reasonable in view of the large metastable collector. See R. F. Stebbing, Proc. Roy. Soc. (London) **A241**, 270 (1957).

[21] H. S. W. Massey and C. B. O. Mohr, Proc. Roy. Soc. (London) **A140**, 613 (1933). The cross sections calculated for 200 ev are almost exactly in the ratio of n^{-3}. Comparison with experiment shows the calculated values at 100 ev to be too large relative to those at 200 ev.

[22] D. R. Bates and A. Damgaard, Trans. Roy. Soc. (London) **A212**, 101 (1949).

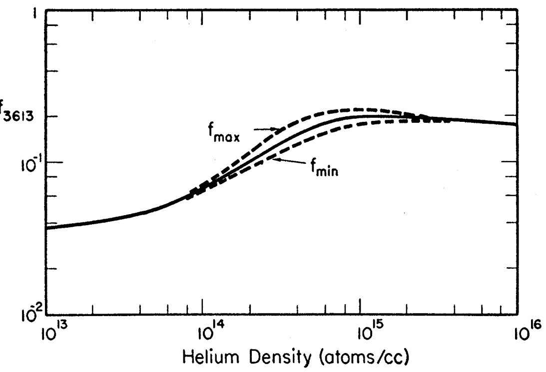

FIG. 7. Theoretical curves showing the effects of imprisonment and excitation transfer on the variation with gas density of the fraction of helium atoms in the $5\,^1P$ state which emit the 3613A line. This calculation was based on an effective radius of 1 cm.

contributions of the various $n\,^1P$ states, the relatively high sensitivity with which metastables can be detected makes measurements possible at low gas densities where the optical experiments fail.

As an illustration of the application of our theory to a case in which the effects of the transfer of excitation are large, we have calculated the apparent cross section for the production of 3613 A radiation by helium atoms in the $5\,^1P$ state. For the purposes of this illustration we make the simplification that all of the excitation transfer is to the $5\,^1D$ state and that cascading effects are negligible. For these assumptions Eq. (5) becomes

$$p_P(\mathbf{r})+\gamma\int n(\mathbf{r}')G(|\mathbf{r}-\mathbf{r}'|)dv'-\gamma n(\mathbf{r}) -A_Pn(\mathbf{r})-\alpha Nn(\mathbf{r})+\alpha' Nm(\mathbf{r})=0, \quad (16)$$

where α is the average value of the product of velocity of the atoms and the cross section for transfer from the $5\,^1P$ state to the $5\,^1D$ state, α' is the average velocity times cross section for transfer from the $5\,^1D$ state to the $5\,^1P$ state, and $n(\mathbf{r})$ and $m(\mathbf{r})$ are the densities of atoms in the $5\,^1P$ and $5\,^1D$ states. The equation for the $5\,^1D$ atoms is

$$p_D(\mathbf{r})-A_Dm(\mathbf{r})-\alpha' Nm(\mathbf{r})+\alpha Nn(\mathbf{r})=0. \quad (17)$$

Eliminating $m(\mathbf{r})$, we obtain

$$p_P(\mathbf{r})+\beta p_D(\mathbf{r})+\gamma\int n(\mathbf{r}')G(|\mathbf{r}-\mathbf{r}'|)dv' -[\gamma+A_P+(1-\beta)\alpha N]n(\mathbf{r})=0, \quad (18)$$

where $\beta=\alpha' N/(A_D+\alpha' N)$. Since cascading is assumed negligible, the two production terms have the same spatial distribution and Eq. (18) can be solved by use of the same procedure as for Eq. (5). Figure 7 shows the value of f_{3613} calculated by using the average value of $s(k_0R)$ and $g(k_0R)$ to replace the imprisonment terms in Eq. (18). The important thing to note from this curve is that the apparent cross section for the production of the 3613 A radiation at pressures of the order of that used by Lees ($N=1.5\times10^{15}$ atoms/cc) is never more than 20% of the total cross section for the $5\,^1P$ and $5\,^1D$ states and is almost 6 times the value which would be observed if there were no imprisonment. In this calculation we assumed an excitation transfer cross section approximately equal to the sum of the observed $5\,^1P-5\,^1D$ and $5\,^1P-5\,^3D$ cross sections,[17] i.e., 10^{-13} cm^2. If the $5\,^3D$, $5\,^1F$, and $5\,^3F$ states had been included in the analysis, the curve of f_{3613} *vs* N would have been much the same at low and intermediate densities but would have dropped to a somewhat lower value at the highest densities. This example shows that it is necessary to take into account both imprisonment and the transfer of excitation when evaluating the measured cross sections. The cross section for the excitation of the $5\,^1P$ state, when one uses Lees' data multiplied by $3/f$, is found to be 8.5×10^{-19} cm^2. This value is to be compared to the theoretical cross section[21] of 8×10^{-19} cm^2 and the value of 5×10^{-19} cm^2 obtained from our analysis of Dorrestein's data for 100-ev electrons.

VI. DISCUSSION

The analysis presented in this paper shows that in order to obtain significant values for the cross section for the production of atoms in a state which emits both resonance and nonresonance radiation, one must take into account the imprisonment of the resonance radiation. In particular we have shown that when properly analyzed, the published experimental data are in reasonable agreement with the predictions of theory. We have shown that if experiments are made in sufficiently ideal geometry, one should be able to make quantitative comparisons between the theoretical and experimental variations of the apparent cross sections with gas density. The available data show as good agreement as can be expected in view of the nonideal experimental conditions. Obviously, further experiments are required.

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