Total Cross Sections for Inelastic Scattering of Charged Particles by Atoms and Molecules. III. Accurate Bethe Cross Section for Ionization of Helium†

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The Bethe cross section σ_i for ionization of He by fast charged particles is accurately evaluated by a subtraction $\sigma_i = \sigma_{tot} - \sigma_{ex}$, where σ_{tot} is the total inelastic scattering cross section and σ_{ex} is the sum of all discrete-excitation cross sections. Our earlier work has given a highly precise value of σ_{tot}, and recent results on discrete excitations enables one to determine σ_{ex}. The resulting "counting" ionization cross section for a particle of charge ze and velocity $v = \beta c$ is

$$\sigma_i = \frac{8\pi a_0^2 z^2}{mv^2/R}\left\{0.489\left[\ln\left(\frac{\beta^2}{1-\beta^2}\right) - \beta^2\right] + 5.526\right\},$$

where m is the electron mass, a_0 is the Bohr radius, and R is the Rydberg energy. Among numerous measurements, the electron-impact data by Smith are most consistent with our result and suggest a gradual attainment of the Bethe asymptotic behavior near 1-keV incident electron energy.

I. INTRODUCTION

The first Born approximation should give accurately the cross section for excitation or ionization of an atom or molecule by sufficiently fast charged particles.[1] However, serious attempts to calculate accurate cross sections usually encounter the difficulty of obtaining reliable wave functions of the atom or molecule, especially wave functions for an ionization continuum. The many published calculations on the ionization of He all utilize approximate, often crude, wave functions[2] which introduce uncertainties that are, in general, hard to estimate.

This difficulty is circumvented by the use of the sum rule for the Bethe cross sections[3]; knowing the total inelastic scattering cross section σ_{tot} obtained thereby, one can write the ionization cross section σ_i as

$$\sigma_i = \sigma_{tot} - \sigma_{ex}, \tag{1}$$

where σ_{ex} is the sum of the cross sections over all discrete excitations. This procedure, in principle applicable to any atom or molecule, is illustrated below in the case of He; accurate Bethe cross sections for several most important discrete excitations have recently become available,[4-6] and make possible a reliable determination of σ_{ex}.

II. CALCULATION

The Bethe cross section σ_s for an optically allowed transition to the state s by a particle of charge ze and velocity v is[1,3,4]

$$\sigma_s = 4\pi a_0^2 z^2 (R/T) M_s^2 \ln(4c_s T/R), \tag{2}$$

where a_0 is the Bohr radius, R is the Rydberg energy and $T = \frac{1}{2}mv^2$, m being the electron mass. Further, M_s^2 is the dipole-matrix-element squared and is equal to Rf_s/E_s, where f_s is the dipole oscillator strength and E_s is the energy for excitation to the state s. The parameter c_s is defined by

$$\ln\left[c_s\left(\frac{E_s}{R}\right)^2\right] = \int_0^\infty \frac{f_s(K)}{f_s} d\ln(Ka_0)^2 - \int_{-\infty}^0 \left[1 - \frac{f_s(K)}{f_s}\right] d\ln(Ka_0)^2, \tag{3}$$

where $f_s(K)$ is the generalized oscillator strength at momentum transfer $\vec{K}\hbar$. For an optically forbidden transition to the state s', the corresponding expression is [1,3,4]

$$\sigma_{s'} = 4\pi a_0^2 z^2 (R/T) b_{s'}, \tag{4}$$

where $$b_{s'} = \int_{-\infty}^\infty \frac{f_{s'}(K)}{E_{s'}/R} d\ln(Ka_0)^2 . \tag{5}$$

The summation of the Bethe cross sections of the forms (2) and (4), over all discrete transitions, gives

$$\sigma_{ex} = 4\pi a_0^2 z^2 (R/T) M_{ex}^2 \ln(4c_{ex} T/R) , \quad (6)$$

where $$M_{ex}^2 = \sum\nolimits_{\text{discrete}} M_s^2 \quad (7)$$

and $$M_{ex}^2 \ln c_{ex} = \sum\nolimits_{\text{discrete}} (M_s^2 \ln c_s + b_{s'}). \quad (8)$$

Accurate theoretical values of the parameters M_S^2, $\ln c_S$, and $b_{S'}$ are available for $n^1P(n=2-4)$, $n^1s(n=2-7)$, and 3^1D excitations,[4-6] and also serve as a reliable basis for extrapolating the parameters for other excitations. Table I summarizes the data thus collected.[7] The summations in Eqs. (7) and (8) thus yield

$$M_{ex}^2 = 0.2633 \quad (9)$$

and $$M_{ex}^2 \ln c_{ex} = -0.4164. \quad (10)$$

Although the data of Table I are judged to be highly trustworthy on theoretical grounds alone, their compatibility with pertinent experimental information has been tested in a variety of ways. One example concerns the leading terms (n^{-3}) of the extrapolation formulas in Table I. Assuming smooth variation of the generalized oscillator strength with respect to excitation energy at the (first) ionization threshold I, one may write

$$\lim_{n\to\infty} \tfrac{1}{2} n^3 f_n(K) = R \left. \frac{df(K,E)}{dE} \right|_{E=I} , \quad (11)$$

where $df(K,E)/dE$ is the differential generalized oscillator strength for the transition into continua at excitation energy E. The use of Eq. (11), together with Eqs. (3) and (5) leads to

$$\lim_{n\to\infty} \tfrac{1}{2} n^3 (M_{n^1P}^2 \ln c_{n^1P} + b_{n^1S} + b_{n^1D} + \cdots)$$

$$= \frac{R^2}{E} \frac{df}{dE} \ln c_E \Big|_{E=I} , \quad (12)$$

where df/dE is the differential dipole oscillator strength and $\ln c_E$ is defined by

$$\ln\left[c_E \left(\frac{E}{R}\right)^2\right] = \int_0^\infty \frac{df(K,E)/dE}{df/dE} \, d\ln(Ka_0)^2 - \int_{-\infty}^0 \left[1 - \frac{df(K,E)/dE}{df/dE}\right] d\ln(Ka_0)^2, \quad (13)$$

The evaluation of the left-hand side of Eq. (12) by use of the data in Table I results in a value of -0.74. The right-hand side of Eq. (12) can be evaluated from the experimental data of Lassettre *et al.*[8] Because the measurements of Ref. 8 cover only a rather limited range of K, calculation of the integrals in Eq. (13) requires extrapolation of the data. To this end, it appears best to use a fitting formula as suggested by Lassettre[9]

$$\frac{df(K,E)}{dE} = \frac{df}{dE} \frac{1}{(1+x)^6} \sum_{\nu=0}^{N} g_\nu \left(\frac{x}{1+x}\right)^\nu , \quad (14)$$

TABLE I. Parameters for the Bethe cross sections for discrete excitation of He and their sums.

n	Allowed transition to the n^1P states M_n^2		$M_n^2 \ln c_n$
2	0.17708[a]		−0.3307
3	0.0433[a]		−0.0793
4	0.0173		−0.0314
$n \geqq 5$[b]	$1.01\,(n^*)^{-3} + 1.85\,(n^*)^{-5}$		$-1.85\,(n^*)^{-3} - 2.82\,(n^*)^{-5}$
sum	0.2633		−0.4880
	Forbidden Transitions[c]		
n	$b(n^1S)$	n	$b(n^1D)$
2	0.0455		
3	0.0103	3	0.0025
4	0.0039	$n \geqq 4$[b]	$0.104\,(n^*)^{-3} - 0.33\,(n^*)^{-5}$
$n \geqq 5$[b]	$0.209\,(n^*)^{-3} + 0.24\,(n^*)^{-5}$		
sum	0.0653	sum	0.0062

[a]Taken from B. Schiff and C. L. Pekeris, Phys. Rev. 134, A638 (1964).
[b]In the extrapolation formulas, $n^* = n + \delta$, where $\delta = 0.0121$, -0.140, and -0.00209 for ^1P, ^1S, and ^1D, respectively [M. J. Seaton, Proc. Phys. Soc. (London) 88, 815 (1966)].
[c]The $b_{S'}$ for all the n^1F and the other excitations is estimated to be $0.05n^{-3} - 1.2n^{-5}$, leading to the sum of ~ 0.0001 (Ref. 7).

where the g_ν are constants and $x=(Ka_0)^2R/I$. Using Eq. (14) with $N=3$, 4, and 5, we conclude that the data of Ref. 8 yield the value -0.73 ± 0.04 for the right-hand side of Eq. (12), in satisfactory corroboration of the value from Table I.

The substitution of σ_{ex} [Eqs. (6)–(10)] and of the accurately known σ_{tot} into Eq. (1) leads to[3,10]

$$\sigma_i = 4\pi a_0^2 z^2 (R/T) M_i^2 \ln(4c_i T/R), \qquad (15)$$

with
$$M_i^2 = 0.489, \qquad (16)$$

and
$$M_i^2 \ln c_i = 0.036, \qquad (17)$$

where (16) is the total dipole-matrix-element squared for ionization.[11,12] An estimate of the numerical uncertainties in Eqs. (16) and (17) indicates that the above σ_i should be accurate, for instance, within the framework of the Bethe theory, to 1% or better for $T \geqq 1$ keV. A modification by relativistic kinematics is required for $T \geqq 10$ keV, and the result is then[3,4,11]

$$\sigma_i = \frac{8\pi a_0^2 z^2}{mv^2/R}\{0.489[\ln\left(\frac{\beta^2}{1-\beta^2}\right) - \beta^2] + 5.526\}, \qquad (18)$$

where $\beta = v/c$.

III. CONCLUSION

The Bethe cross sections σ_{tot}, σ_i, and σ_{ex} are shown in Fig. 1, where the ordinate represents $(T/R)\sigma/4\pi a_0^2 z^2$ and the abscissa represents $\ln(T/R)$, a plot that was first suggested by Fano.[11] Experimental data on the ionization[13-19] are also included for comparison.

Owing to the nature of our theory, the σ_i [Eqs. (15) or (18)] corresponds to the simple sum of the single- and double-ionization cross sections, which sum is sometimes called the "counting" ionization cross section. For He, however, its difference from the "gross" ionization cross section as determined by a total-current measurement, is barely significant in the following comparison with experiment, because the double-ionization cross section is very small.[14,15]

Of the many electron-impact experiments, the data by Smith[13] are consistent with theory and indicate a gradual attainment of the asymptotic behavior around $T \cong 1$ keV. The data of Schram *et al.*[14] and of Gaudin and Hagemann,[15] are incompatible with theory in spite of the apparently reasonable asymptotic slopes seen in Fig. 1. Actually, the known values of M_{ex}^2 and M_i^2[Eqs. (9) and (16)] alone suggest the difficulty of reconciling these two sets of the ionization data with the sum rule of Ref. 3, which one may express as

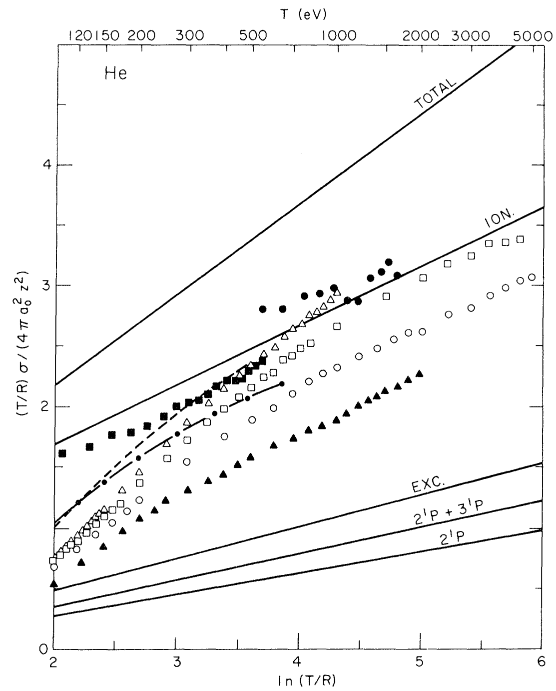

FIG. 1. Straight lines represent, respectively, from top to bottom, the Bethe cross sections for total inelastic scattering (TOTAL), for ionization (ION.), for excitations to all discrete levels (EXC.), for excitations to the 2^1P and 3^1P levels, and for the 2^1P excitation only. The ionization cross section is to be compared with the following data. The open squares show the electron-impact data by Smith (Ref. 13), the open circles are those by Schram *et al.* (Ref. 14), the closed triangles are those by Gaudin and Hagemann (Ref. 15), and the open triangles are those by Rapp and Englander-Golden (Ref. 16). The closed squares show the proton-impact data by Hooper *et al.* (Ref. 18), and the closed circles show those by Pivovar and Levchenko (Ref. 19). The dashed line represents the calculation by I. H. Sloan [Proc. Phys. Soc. (London) 85, 435 (1965)], and the dot-dashed line represents that by G. Peach [Proc. Phys. Soc. (London) 85, 709 (1965)].

$$M_{ex}^2 \ln c_{ex} + M_i^2 \ln c_i = M_{tot}^2 \ln c_{tot} = -0.380. \qquad (19)$$

Let $\ln c_E$ denote the constant in the Bethe cross section of form (2) for excitation to all the states at excitation energy E, either in the discrete or the continuous spectrum. The dependence of $\ln c_E$ on E is a characteristic of the Bethe surface,[20] gross features of which should be carried over from H to He (double excitations in the latter being disregarded). Then, for He, it is reasonable to anticipate that $\ln c_E$ is an increasing function of E, as is the case for H.[21] Since $\ln c_{ex}$ and

$\ln c_i$ are simply averages of $\ln c_E$ weighted with the square of the dipole matrix element, one then expects $\ln c_i > \ln c_{ex}$, and thence [from Eqs. (9), (16), and (19)] $\ln c_i > -0.505 > \ln c_{ex}$. The data of Refs. 14 and 15, however, correspond to $\ln c_i$ values of about -0.99 and -1.99, respectively. This difficulty with reconciling the data of Ref. 14 was noted earlier by Vriens.[22] (One should use caution, however, in generalizing the above argument to atoms with many shells.)

A comparison with the data by Rapp and Englander-Golden[16] must remain somewhat inconclusive, because their data are limited to $T \leqq 1$ keV, although the magnitude at $T = 1$ keV is close to the theoretical value. The data of Harrison,[17] which are not included in Fig. 1, lie between the data of Smith[13] and those of Schram *et al.*[14] The proton-impact data, for which no high precision is claimed,[18,19] appear reasonably consistent with theory within the experimental uncertainties.

Additional remarks concern earlier theoretical or semiempirical studies, primarily those dealing with electron impact. The ionization cross section adopted by Miller,[23] after evaluation of data then available is in good agreement with our determination. The σ_i deduced by Vriens[22] through an analysis which is similar to ours, but which relies heavily on experimental data on discrete excitations, is now confirmed by the present result on better theoretical grounds. Most of the published calculations[2] explicitly using approximate continuum wave functions present results only for $T \lesssim 500$ eV, where it is uncertain that the Bethe asymptotic behavior is attained. By use of approximate wave functions, however, Perlman[24] evaluated σ_i as well as σ_{tot} in the Bethe asymptotic form, through an approach which is in part similar to our formulation. For example, at $T \cong 1$ keV, the σ_i in Ref. 24 is larger by about 3% and the σ_{tot} is smaller by about 9% than our results. Further, Van de Walle and Grosjean[25] and also Gaudin and Botter[26] give asymptotic formulas which predict a cross section for single ionization that is smaller than our σ_i by 3–5% for $T \cong 1$ keV. Aside from all these studies on electron impact, calculation for any fast charged particles may be compared with our theory, insofar as the particles are regarded as structureless. For example, the calculation by Mapleton[27] for proton impact gives an ionization cross section smaller than our σ_i, again by about 5% at 2-MeV proton energy (or $T \sim 1$ keV). (Note that the percentage differences depend on T.)

In conclusion, the inconsistency among different sets of experimental data suggests that sizable systematic errors are still present in current measurements.[28] We hope that our determination of the asymptotic σ_i will stimulate an improved experiment in the future. The apparent close agreement of Smith's data[13] with our calculation may well be in part accidental, inasmuch as studies conducted since 1930 have uncovered additional sources of systematic errors, which were probably unknown at that time.[28] Besides, whereas our value for σ_i is certainly accurate asymptotically, one has at present no dependable means for theoretically determining the velocity at which the Bethe asymptotic behavior is attained; one must continue to consult experimental work on this important question.

Note added in proof. Recently, K. L. Bell and A. E. Kingston [J. Phys. B2, 653 (1969)] calculated the ionization cross section of He by proton impact, using a six-parameter correlated ground-state and partial-wave ($l \leqq 5$) continuum wave functions. Their cross section is 1 ~ 3% larger than our result in the asymptotic region.

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$I_1 = 1.0811$, $I_2 = 0.1851$, and thence $M_{tot}^2 \ln c_{tot} = -0.3804 \pm 0.0040$. This confirms the result of Ref. 3. A tabulation of the atomic form factor and the incoherent-scattering function computed from the Weiss wave function is available from the authors upon request.

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