Relative and absolute level populations in beam-foil–excited neutral helium*

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Relative and absolute populations of 19 levels in beam-foil–excited neutral helium at 0.275 MeV have been measured. The singlet angular-momentum sequences show dependences on principal quantum number consistent with n^{-3}, but the triplet sequences do not. Singlet and triplet angular-momentum sequences show similar dependences on level excitation energy. Excitation functions for six representative levels were measured in the range 0.160 to 0.500 MeV. The absolute level populations increase with energy, whereas the neutral fraction of the beam decreases with energy. Further, the P angular-momentum levels are found to be overpopulated with respect to the S and D levels. The overpopulation decreases with increasing principal quantum number.

I. INTRODUCTION

In recent years, beam-foil spectroscopy has become a valuable experimental method of atomic physics. However, relatively little attention has been devoted to the beam-foil excitation process itself. The beam-foil literature contains many general, often contradictory comments about the interaction, but few results from systematic approaches to the problem. A series of absolute and relative quantum-level population measurements would be of great value to an understanding of the interaction.

At present no theory exists for excited-state production using the beam-foil method. A number of existing theories may apply, however. These fall into three rather broad categories: those adopted from atom-atom collisions, those adopted from recombination at the surface of a metal, and those we shall call "others."

Among those adopted from atomic-collision theory is radiative capture of free electrons by bare nuclei in the high-velocity limit.[1] This approach leads to an n^{-3} dependence on principal quantum number of the capture cross section. However, Lennard and Cocke[2] have suggested that the probabilities for this process are too small to account for their experimental results.

Also in this category are a number of Born-approximation calculations for the capture of bound electrons by fast projectiles.[3-6] These also have in common an n^{-3} dependence of the cross sections on principal quantum number. Bates and Dalgarno[7] have calculated cross sections for capture into various angular-momentum levels for the $1s$ state of hydrogen. At high energies (250 keV/amu) they find the P states to be more populated than S states, which are more populated than D states, and the principal-quantum-number dependence is approximately n^{-3}. At 10 keV/amu, the cross sections diverge from the n^{-3} dependence.

Hiskes[8] has done similar calculations, but has taken into account the effects of targets different from hydrogen. His results also show an n^{-3} dependence, which in some cases is obeyed even for small energies and small n.

Trubnikov and Yavlinskii[9] have considered the case of tunnel recombination near the surface of a metal foil. They also find that the relative excited-state production goes as n^{-3}. McLelland,[10] however, believes that their assumptions make their results inapplicable to nonmetals. He considers a model applicable to semimetals such as graphite, and also reports an n^{-3} dependence.

In the category we call "other" are rather general ideas which are incapable of predicting an actual n dependence, but which locate most of the population in some level.

If we adopt the suggestion of Bohr[11] that the favored states in a capture are those for which the electron velocity is close to the ion velocity, we find most of the population to be in states with quantum number $n \approx (Q+1)[24.8A_{\text{ion}}/E_{\text{ion}}(\text{keV})]^{1/2}$.[12] Here, Q is the ionic charge, A_{ion} is the atomic weight, and E_{ion} is the incident ion energy.

According the Oliphant and Moon,[13] capture is favored into states bound with an amount of energy close to the work function of the surface. For carbon this yields states with quantum number $n \approx 1.7(Q+1)$.[12]

There is little beam-foil data in existence to compare with these theories. Most previous relative-intensity measurements have been limited to levels having transitions with closely spaced wavelengths,[14,15] which eliminates the problem of calibrating the spectrometer over a wide wavelength range. Andersen *et al.*[16] used a calibrated system to do a study at 40 keV similar to ours. Their work, however, suffers from inadequate normalization procedures and lack of attention to polarization problems. Lennard and Cocke[2] used a calibrated system to measure absolute excitation probabilities for hydrogenlike states in iron. Their data show an approximate n^{-3} dependence but are limited to high-n states of high-charge states.

We calibrated an experimental station and measured both relative and absolute populations of low-lying levels ($3 \leq n \leq 6$) in neutral helium. At 0.275 MeV a fair amount of neutral helium is produced by the beam-foil source.[17] Neutral helium has roughly 20 transitions from a good range of upper-level quantum numbers in the visible and near ultraviolet. In this region the system may be easily calibrated with a black-body standard lamp.

II. DESCRIPTION OF EXPERIMENT

The experiment is divided into two parts. Initially, the entire system must be calibrated as a function of wavelength to give absolute numbers of photons. Next, the photons from the beam are counted and corrected by geometric and dynamic quantities to give photons per emergent neutral helium atom.

The experimental arrangement is shown in Fig. 1. Light from either the standard source or the beam-foil source enters an achromatic optical system which was designed such that the optical paths from the two sources be virtually identical. For this purpose, the concave mirror may be rotated to accept light from either source, and focus it with magnification one at the entrance slit of spectrometer #1.

The light then passes through a wedge interference filter which reduces the scattered light present during use of the standard lamp, and which remains in place during beam measurements to preserve the system efficiency identically. Spectrometer #1 (Heathkit) selects the wavelength of interest, and the phototube (EMI 6256S) acts as a photon counter. Spectrometer #2 (Heathkit) is used for normalization purposes; it monitors the number of photons of one helium line (λ 4713 Å).

A. Calibration of system

The entire optical and electronic portion of the experimental system was calibrated as a function of wavelength. The absolute efficiency $\epsilon(\lambda)$ in counts per photon between λ 2800 and λ 6000 Å was obtained using a General Electric incandescent tungsten-ribbon filament lamp, with its intensity calibration traceable to the National Bureau of Standards.

The entrance- and exit-slit widths of spectrometer #1 were calibrated using the diffraction of laser light. Since different slit widths were used for the calibration and beam measurements, the linearity of intensity from a line source as a function of slit width was demonstrated. The instrumental polarization of the optical system was measured and used in correcting the number of photons counted from the beam. It was shown that the standard-lamp polarization was less than 1% throughout the spectral range of interest.

To obtain the efficiency $\epsilon(\lambda)$, one must calculate

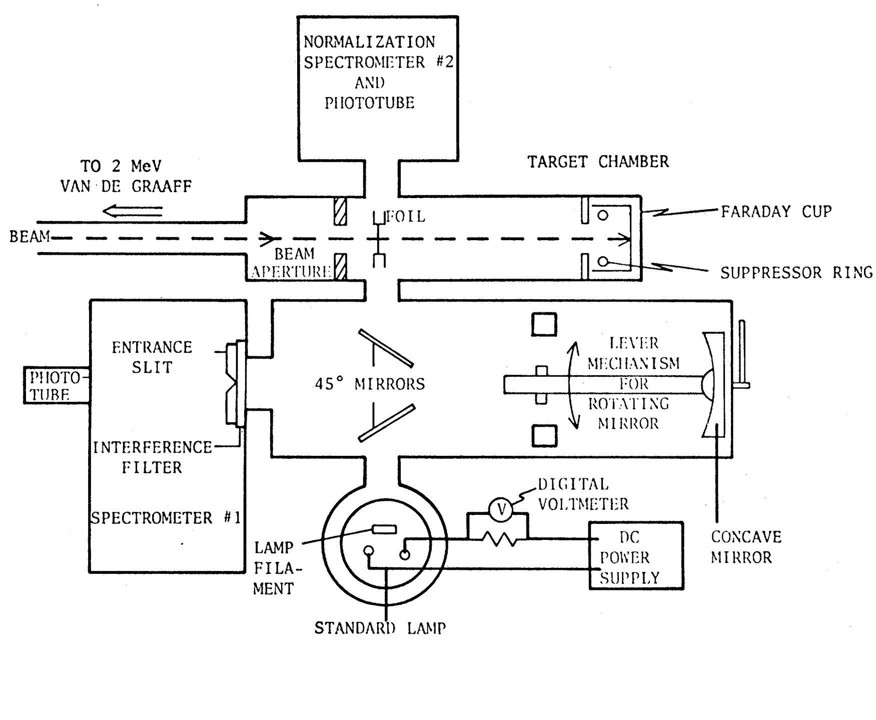

FIG. 1. Experimental station.

from Planck's law the number of photons per second, N, emitted by the standard lamp, and observe the counts per second, C, recorded by the experimental system:

$$N = N[E(\lambda, T), \tau_g(\lambda), L_s, W_s, M, \Delta\Omega, D_l(\lambda), T, \lambda],$$

where $E(\lambda, T)$ is the emissivity of tungsten, $\tau_g(\lambda)$ the transmission of lamp window, L_s and W_s the length and width of spectrometer slits, M the system magnification, $\Delta\Omega$ the solid acceptance angle, $D_l(\lambda)$ the linear dispersion of the spectrometer, T the absolute temperature of standard lamp, and λ the wavelength. Numbers of photons were calculated taking accurate account of all these parameters, and absolute and relative efficiencies were obtained for more than fifty wavelengths between λ 2800 Å and λ 6000 Å.

The uncertainty in the absolute efficiency was calculated taking into account the individual uncertainties in all of the above parameters as well as the statistical uncertainty resulting from the observed count rate. The absolute uncertainty is wavelength dependent, and varies from 32% at λ 3819 Å to 20% at λ 5875 Å, and is dominated by the uncertainty in the temperature of the standard lamp.

The statistical uncertainties involved in the calibration are much smaller than the overall uncertainty, hence the measured relative light intensities are more accurate than the absolute intensities. The uncertainty in the relative efficiency is less than 10% for all wavelengths of interest above λ 3447 Å. The uncertainties in the relative efficiencies are larger below λ 3800 Å due to the presence of scattered light. Since the interference filter did not transmit in this region, the scattered-light intensity was measured for wavelengths below the spectral range of the standard lamp, and observed helium line intensities in the region λ 2800 Å–λ 3800 Å were corrected by this amount.

B. Measurement of relative and absolute level populations of neutral helium

Helium ions were accelerated to 0.275 MeV by a 2-MeV Van de Graaff and passed through a 6-μg/cm^2 carbon foil. Light from levels of interest was observed at a distance 3 mm downstream from the foil at 90° to the beam. The target-chamber geometry and the solid acceptance angle of the optical system prevented viewing the beam at the foil directly; hence, the smallest possible distance downstream (3 mm) was used. For this distance, cascade contributions were calculated based on the conservative assumption that all levels are equally populated. It was found that the cascade contribution to the measured populations was less than 2% in all cases at 3 mm downstream.

The target chamber was equipped with a shielded, suppressed Faraday cup (aperture 1 in.) located 4.0 in. downstream from the foil. The size of the beam was determined by a 3-mm-square beam aperture 1 cm upstream from the foil. Estimates of the rms scattering angle for helium in 6-μg/cm^2 carbon at 275 keV showed that scattering had virtually no effect on normalization procedures.

Two methods of data normalization were used. For charge normalization, the current from the Faraday cup was integrated, and the light per unit beam charge was counted. The neutral fraction of the beam was calculated using a charge-state distribution measurement by Bickel.[17] From this, the number of photons per emergent neutral helium atom was calculated.

For spectral-line-intensity normalization, a second monochromator monitored λ 4713 Å , and the light per unit λ 4713 Å was counted for a preselected time determined by the approximate relative intensity of the spectral line. All measurements were normalized to a fixed number of counts of λ 4713 Å, this being sufficient to obtain relative populations. It was observed that the spectral-line-intensity normalization method gave more reproducible results for relative populations, hence this method was used for all data taken at 0.275 MeV.

Since beam radiation is polarized,[18] the polarizations of all spectral lines were measured. The correction to the level populations from the anisotropy of the radiation and instrumental polarization effects was small in all cases, never exceeding 4%.

The system was first calibrated using the standard lamp. The calibration procedure was repeated many times over a period of several weeks. The relative calibration was found to be reproducible to less than 10% in all cases. Deviations greater than this were indicative of system malfunctions.

Before and after each beam measurement, the system calibration was checked. A spectral line was selected, and the interference filter was adjusted to give maximum transmission at this wavelength. Counts were recorded for both monochromators, and all measurements were normalized.

From the observed light intensity and the system efficiency $\epsilon(\lambda)$, the number of atoms in a level n, l, S per emergent neutral atom was calculated. Target-chamber geometry, solid acceptance angles, exponential decay downstream, the precise volume of the beam observed, and polarization effects, were accurately taken into account.

III. RESULTS

A. Dependence of populations on principal quantum number

The measured relative and absolute level populations are given in Table I. Populations of the $ns\,^1S$, $np\,^1P$, $nd\,^1D$, $ns\,^3S$, and $nd\,^3D$ sequences were plotted against n^{-1} on log-log plots, and least-square fits were done. Two representative plots are given in Figs. 2 and 3. The resulting best-fit equations are given in Table II. The singlet sequences show population dependences consistent with n^{-3}; the triplets, however, do not.

As Andersen and co-workers[16] have pointed out, the data may also be fitted by an exponential equation of the form $C_1 e^{-C_2 n}$, where C_1 and C_2 are constants. There is no supporting theory for this dependence, which seems to be a consequence of the small range of quantum levels observed. Observation of transitions from levels $n=2$ or $n>6$ would distinguish between the two. We observed the $(7d\,^3D \rightarrow 2p\,^3P)$ λ 3705 Å transition, and measured the population of the $7d\,^3D$ level. The result, plotted with the rest of the data for the $nd\,^3D$ sequence and the two least-square fits (Fig. 4), shows strong but not conclusive evidence for the n^{-3} type dependence.

The measured relative and absolute populations have a number of consequences.

(1) We did not measure the populations of levels with $n=1$ and $n=2$. If the behavior we have found for $n \geq 3$ can be extrapolated to these lower levels, we find that the ground state is not greatly overpopulated with respect to the next few higher-lying levels. For the case of neutral helium, this tends to discount the theories we have called "other."

(2) The question of whether the populations are closed has occurred in the literature.[19] If the populations fall off as fast as n^{-2} or n^{-3}, then the total absolute population converges as expected.

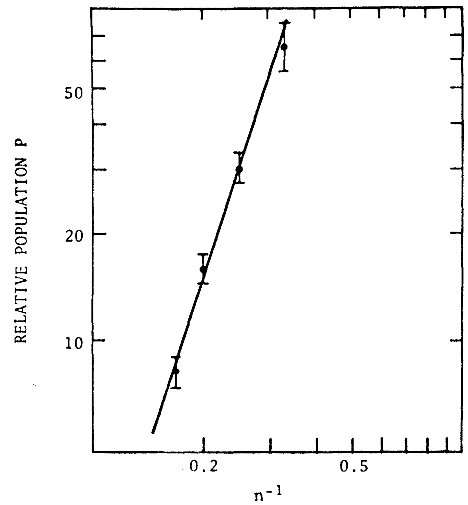

FIG. 2. Relative population of $np\,^1P$ sequences as a function of n^{-1} at 0.275 MeV. The line represents the least-square-fit equation $\log P = 14.5(0.4) + 3.0(0.2)\log n^{-1}$.

TABLE I. Relative and absolute populations. (The numbers in parentheses are the percentage uncertainties.)

λ (Å)	Transition	Relative population of upper level (No. excited atoms/ emergent neutral)	Absolute population of upper level (No. excited atoms/ emergent neutral $\times 10^{-3}$)
3187	$2s\,^3S - 4p\,^3P$	165.0 (20)	16.2 (51)
3447	$2s\,^1S - 6p\,^1P$	8.2 (10)	0.8 (42)
3613	$2s\,^1S - 5p\,^1P$	16.1 (9)	1.6 (41)
3819	$2p\,^3P - 6d\,^3D$	15.2 (13)	1.5 (44)
3867	$2p\,^3P - 6s\,^3S$	12.2 (23)	1.2 (46)
3889	$2s\,^3S - 3p\,^3P$	258.0 (9)	25.4 (41)
3964	$2s\,^1S - 4p\,^1P$	30.3 (10)	3.0 (41)
4026	$2s\,^3S - 5d\,^3D$	27.8 (8)	2.7 (40)
4121	$2p\,^3P - 5s\,^3S$	19.9 (8)	2.0 (39)
4144	$2p\,^1P - 6d\,^1D$	6.0 (13)	0.6 (41)
4169	$2p\,^1P - 6s\,^1S$	6.1 (15)	0.6 (41)
4389	$2p\,^1P - 5d\,^1D$	10.2 (15)	1.0 (41)
4437	$2p\,^1P - 5s\,^1S$	8.6 (24)	0.9 (44)
4471	$2p\,^3P - 4d\,^3D$	49.7 (9)	4.9 (38)
4713	$2p\,^3P - 4s\,^3S$	52.0 (10)	5.1 (37)
4922	$2p\,^1P - 4d\,^1D$	19.4 (10)	1.9 (39)
5015	$2s\,^1S - 3p\,^1P$	65.8 (16)	6.5 (38)
5047	$2p\,^1P - 4s\,^1S$	18.4 (18)	1.8 (40)
5875	$2p\,^3P - 3d\,^3D$	104.0 (10)	10.2 (36)

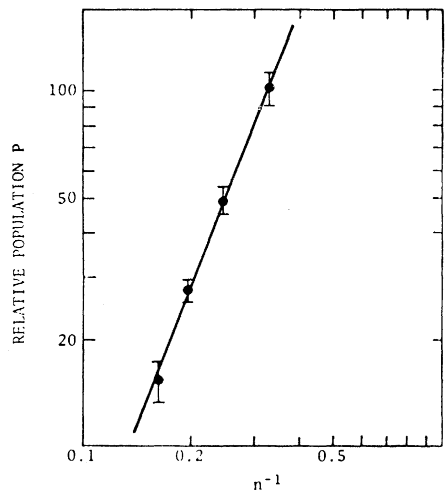

FIG. 3. Relative populations of $nd\,^3D$ sequence as a function of n^{-1} at 0.275 MeV. The line represents the least-square-fit equation $\log P = 14.5(0.3) + 2.7(0.2) \log n^{-1}$.

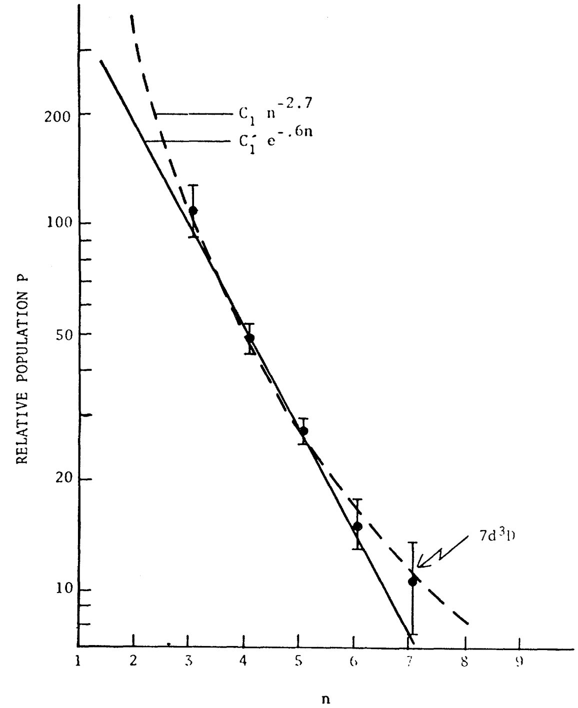

FIG. 4. Relative populations of $nd\,^3D$ sequence at 0.275 MeV as a function of n with additional point. The curves are least-square fits to the data without the additional point.

(3) For cascade analysis in beam-foil mean life studies, it is generally assumed that an infinite number of levels cascade to any given level. An approximate n^{-3} dependence of the populations would indicate that only a few levels immediately above the one in question contribute significantly.

(4) By summing the absolute populations for *all* levels in all angular-momentum sequences we observed, we find that 40%–70% of the populations must be in levels with $l > 2$ or must be multiply excited. Hence a significant fraction of the total population is in the higher angular-momentum levels.

B. Dependence of populations on excitation energy

The relative state populations are shown as functions of level excitation energy in Fig. 5. Clearly a temperature dependence[20] in the form of a Boltzmann distribution is not valid. However, the higher excited levels have a slope which yields a temperature of roughly 6000 °K. The slope of the lower levels yields approximately 9000 °K.

In Fig. 5, the similarity in shape of the curves for sequences with the same orbital angular momentum but different spin is remarkable. This suggests that the populations exhibit systematic behavior, and that the excitation energy may be a useful quantity for a rough but general parametrization of the beam-foil interaction.

C. Excitation functions

The absolute populations as functions of incident-particle energy between 0.16 and 0.50 MeV were measured for six representative levels using the charge-normalization procedure. Polarization varies as a function of energy,[21] but the correction is small (2%) for a relatively large change in polarization (5%–20%). Therefore, this correction was not made.

The results are given in Table III. Log-log

TABLE II. Least-square-fit equations for relative populations as functions of n^{-1}. (The numbers in parentheses are the absolute errors in the quantities.)

l	Singlets	Triplets
S	$\log P = 13.5(0.9) + 2.7(0.6) \log n^{-1}$	$\log P = 16.4(0.8) + 4.0(0.5) \log n^{-1}$
P	$\log P = 14.5(0.4) + 3.0(0.2) \log n^{-1}$	
D	$\log P = 13.9(0.6) + 2.9(0.4) \log n^{-1}$	$\log P = 14.5(0.3) + 2.7(0.2) \log n^{-1}$

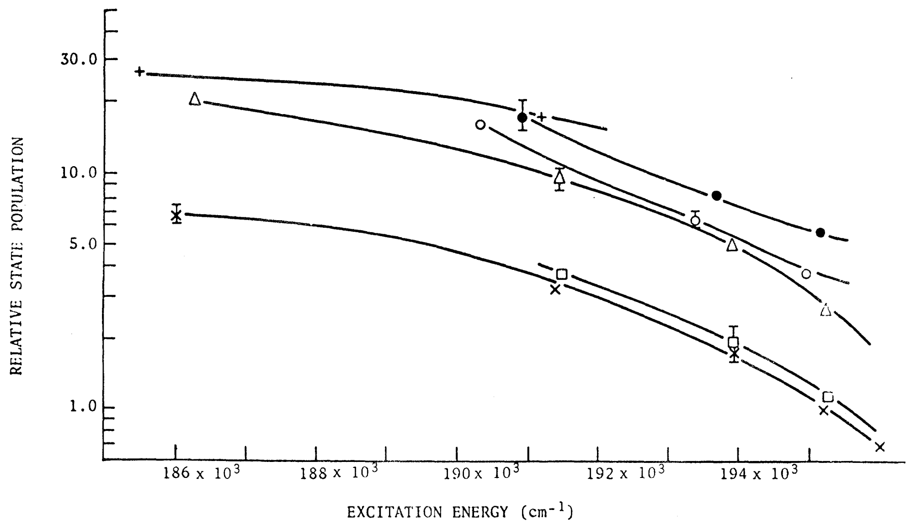

FIG. 5. Relative level populations divided by level statistical weight as functions of level excitation energy. Typical uncertainties are shown. Curves are drawn to guide the eye. ● $ns\,^1S$, △ $np\,^1P$, □ $nd\,^1D$, ○ $ns\,^3S$, + $np\,^3P$, × $nd\,^3D$.

plots of these data show that the populations of the levels we measured are increasing, going roughly as E^k, where k varies from 0.4 to 1.0. Since the total number of neutrals per microcoulomb of charge through the foil is decreasing,[17] some level populations other than those we measured must also be decreasing.

D. Angular momentum dependences

The relative populations are plotted as functions of statistical weights of the levels in Fig. 6. The P levels are overpopulated with respect to the D levels and the S levels, but the overpopulation decreases with increasing n. The singlet populations divided by the statistical weights are shown in Fig. 7. The horizontal line represents the case of purely statistical populations. It appears that the populations become more nearly statistical with increasing n.

The triplet P overpopulation in the $n=4$ level is consistent with the findings of Andersen *et al.*,[16] but the singlet angular-momentum dependences are not. However, Andersen's work was done at 40 keV, and care must be exercised in extrapolating those results to higher energies, since we have found angular-momentum sequences to vary differently with beam energy.

IV. CONCLUSION

We have shown that the relative level populations of beam-foil-excited neutral helium fall off rapidly with increasing n. Singlet dependences are consistent with n^{-3} to within experimental uncertainties whereas the triplet dependences are not. The n dependence shows variation with l and S. The Born-approximation calculations of Bates and Dalgarno[7] and Hiskes[8] predict variations in n dependence with orbital angular momentum; how-

TABLE III. Excitation functions. (The numbers in the table are the absolute populations and are to be multiplied by 10^{-3}. The numbers in parentheses are the percentage uncertainties.)

Energy (MeV)	Level $4s\,^1S$	$3p\,^1P$	$4d\,^1D$	$4s\,^3S$	$3p\,^3P$	$4d\,^3D$
0.160	1.1 (32)	4.8 (30)	1.5 (33)	3.2 (32)	15.8 (29)	4.0 (32)
0.200	1.2 (30)	5.0 (28)	1.4 (31)	3.2 (32)	16.7 (29)	3.8 (31)
0.275	1.6 (31)	5.8 (29)	1.6 (32)	4.5 (32)	22.2 (29)	4.3 (30)
0.300	1.6 (30)	5.5 (30)	1.5 (33)	4.5 (34)	21.9 (29)	4.0 (32)
0.350	1.8 (32)	6.6 (33)	1.9 (39)	5.9 (44)	25.5 (35)	5.0 (41)
0.400	1.8 (49)	7.6 (38)	2.2 (43)	8.6 (45)	30.0 (37)	6.2 (37)
0.500	1.9 (71)	7.6 (45)	2.2 (48)	10.4 (53)	31.7 (45)	6.2 (46)

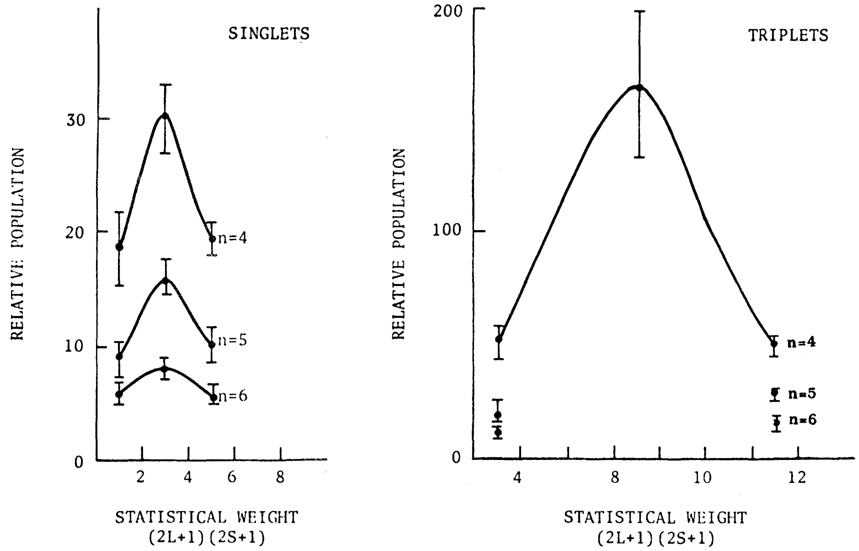

FIG. 6. Relative level populations as functions of the statistical weight of the levels.

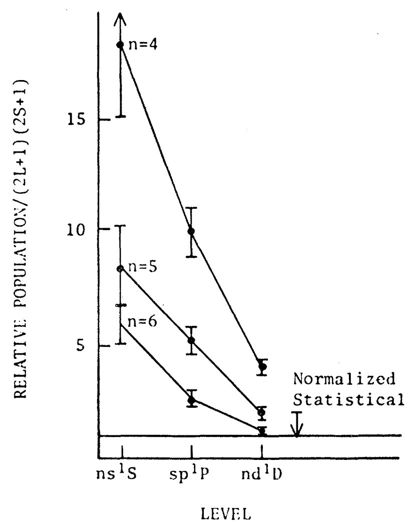

FIG. 7. Relative singlet-level populations divided by the statistical weight of the level. If the populations were strictly proportional to the statistical weights, they would lie on the line marked "normalized statistical."

ever, there is insufficient information at present to make a direct comparison of these theories with our results.

A recent Born-approximation calculation by Omidvar[22] has come to our attention in which the cross sections for production of *ns*, *np*, *nd* states in rearrangement collisions of protons incident on atomic hydrogen are presented. Omidvar finds the *p* states to be overpopulated with respect to the *s* and *d*. For the energy of this experiment, he predicts that the *s* and the *d* populations should be nearly equal. Although these calculations may not be strictly applicable to helium incident on a carbon foil, our results essentially confirm these features.

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