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THE ATOMIC INTERNAL PARTITION FUNCTION

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ABSTRACT

Recent work on the evaluation of the atomic internal partition function at densities less than about 10^{-2} gm cm^{-3} is discussed, in which an attempt is made to identify the physical mechanism responsible for determining the level populations of atoms and ions in dense LTE plasmas. Level populations predicted by a phenomenological theory based on this identification are compared with these obtained by the activity expansion method.

INTRODUCTION

The Boltzmann law for atomic level populations says that the population N_i of state i with energy ϵ_i relative to the lowest state of an atom in thermal equilibrium at temperature T is

$$N_i \sim \exp(-\epsilon_i/kT) \quad . \tag{1}$$

The total number of atoms of the species in question is then proportional to the so-called <u>internal partition function</u>

$$Z(T) \equiv \sum_i \exp(-\epsilon_i/kT) \quad ; \tag{2}$$

degenerate states are represented here by a number of equal terms. As the sum over all bound atomic states includes those near the ionization limit for which $\epsilon_i \to$ const., Z(T) diverges.

This divergence has, of course, been recognized since the earliest days of quantum statistical mechanics. The usual textbook explanation is that interactions with the environment limit the number of bound states, i.e. that as regards the internal state of the particles all gases are non-ideal.

This simple explanation immediately raises three questions, which are not answered. 1) Which interactions are responsible for determining the number of bound states? 2) How many, and which, bound states survive in a given gas or plasma? 3) What <u>are</u> the true level populations of a non-ideal gas? I hope to present some tentative and partial answers here to these questions.

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There are a number of practical applications for such information. The internal partition function must be estimated in calculating ionization equilibria, or more generally, in determining the equation of state of a given gas. Moreover, the level populations must be known to calculate the opacity of the gas. Finally, these considerations play an important role in determining the number of stabilizing states available for dielectronic recombination.

This report is not intended as a comprehensive review of these problems, nor as a complete bibliography of those topics which it does include, but rather as an attempt to present some simple pictures which give insight into the physics involved. First a number of models are discussed that have been employed in the past. The next section mentions briefly a first-principles attack on the problem by means of the activity expansion technique, but concentrates on a phenomenological approach that Dimitri Mihalas and I[1] have employed in connection with an *ab initio* calculation of opacities for stellar envelopes ($\rho < 10^{-2}$ gm cm^{-3}, 3000 K $\leq T \leq 10^7$ K) by an informal international collaboration known as the Opacity Project.[2] In this report I will discuss only the effects of charged particles in determining the level populations.

It is stressed that this is only a progress report on work now under way, and is in no way to be regarded as final. The only conclusion reached here is that experimental guidance is urgently needed in developing an adequate understanding of these phenomena.

HISTORICAL MODELS

Confined Atom Model. This model is based on the reasonable intuitive requirement that a state can not exist if its mean radius is larger than the mean interionic distance r_0, defined by

$$N_{ion} (4\pi/3) r_0^3 \cong 1 \quad . \tag{3}$$

If for simplicity we consider only hydrogenic systems of nuclear charge Z_a, this requirement is approximately

$$n^2 Z_a^{-1} a_0 \lesssim r_0 \quad , \tag{4}$$

where a_0 is the Bohr radius. Thus the principal quantum number of the highest allowed state is

$$n_{max} = (Z_a r_0/a_0)^{1/2} = 1.1 \times 10^4 Z_a^{1/2} N_{ion}^{-1/6} \quad . \tag{5}$$

For hydrogen at a density of $N \sim 10^{18}$ cm^{-3}, we have $n_{max} = 11$.

Static Screened Coulomb Potential (SSCP). In this model, Debye screening converts the Coulomb potential into the SSCP, i.e.

$$V(r) = Z_a/r \tag{6}$$

is replaced everywhere by

$$V(r) = Z_a\, r^{-1}\, e^{-r/D} \quad , \tag{7}$$

where D is the Debye length

$$D \equiv [(4\pi/kT) \sum_j N_j Z_j^2 e^2]^{-1/2} \quad ; \tag{8}$$

for a solar composition $\langle Z^2 \rangle = 2$ and

$$D = 4.9\ (T/N_e)^{1/2}\ \text{cm} \quad . \tag{9}$$

Accurate numerical solutions of the Schrödinger equation have been obtained by several workers; the most complete and accurate are those of Rogers, Graboske and Harwood[3] (RGH). The numerical solutions show that the energy levels are raised relative to the continuum by amounts that depend on the quantum numbers, nuclear charge and Debye length, with the consequence that the number of bound states and therefore Z(T) are finite. RGH give a expression for the maximum allowed value of the principal quantum number,

$$n_{max} = [-\ .1045 + 1.2701\ (DZ_a/a_0)]^{1/2} \quad . \tag{10}$$

From the practical point of view this allows Z(T) to be evaluated in a straightforward manner, although it is disturbing that as D decreases, Z(T) becomes discontinuous every time a bound state "passes into the continuum."

Under what conditions is this model valid? As Debye screening is a consequence of plasma polarization induced by an applied field which acts to cancel that field, we are here concerned with the field induced by the nucleus and the orbital electron. The simplest way to accomplish a partial screening of the nucleus is to require a number of plasma particles to lie within the atomic volume. Thus for a hydrogenic ion of charge Z_a in state n, we must have

$$N_e\ (4\pi/3)\ r_n^3 = N_e\ (4\pi/3)\ a_0^3\ n^6\ Z_a^{-3} \gg 1 \quad . \tag{11}$$

The maximum density at which this is satisfied is approximately

$$N_e^{max}(n) = 1.6 \times 10^{24}\ Z_a^3\ n^{-6} \quad . \tag{12}$$

Thus for the SSCP model to be valid for ground state hydrogen requires solid-state densities. The requirement that the orbital frequency be larger than the plasma frequency, which is necessary for <u>dynamic</u> shielding to occur, also leads to the same conclusion.

A second consideration of principle also disturbs our confidence in the SSCP model. How do we distinguish atomic particles from plasma particles if they are not spatially separated? Not only does the SSCP imply distinguishability, it also requires infinitely fine division of the plasma particles.

Fortunately we can appeal to experiment to support our arguments. The level shifts in a hydrogen plasma with well defined parameters have been measured accurately by Wiese, Kelleher, and Paquette,[4] using a wall-stabilized arc in which $D = 1.6 \times 10^{-6}$ cm. We can calculate the level shifts predicted by SSCP from the accurate energies obtained by RGH. We find:

line:	H_α	H_β	H_γ
exp:	0.55	0.70	0.85 Å
theory:	8.9	7.8	8.3 Å

Thus SCCP appears to overestimate the line shifts by roughly an order of magnitude.

It can be argued that, as the atomic radii are typically very much smaller than the mean interionic distance when $\rho \lesssim 10^{-2}$, the screening of the nuclear potential seen by the orbital electron is caused by image charges within the atomic volume arising from the polarization of the plasma exterior to the atom caused by the "atomic screened" nucleus. Pittman, Voigt and Kelleher[5] show that this effect causes the nuclear charge Z_a appearing in the SSCP to be reduced by unity for a hydrogenic ion, and obtain tolerable agreement with experimental level shifts in the He^+ for $N_e \lesssim 10^{17}$ cm^{-3}. However, this implies that n_{max} is reduced by the factor $[(Z_a-1)/Z_a]^{1/2}$, which leads to the paradoxical result that placing the plasma outside that atom is more effective than having it inside. Moreover, the argument concerning the relative magnitudes of the orbital frequency and the plasma frequency is still valid.

Our conclusion then is that the SSCP is <u>not</u> a valid atomic potential, and in particular does not provide a reliable model for the calculation of atomic level populations and partition functions. This approach fails primarily because the plasma charge is distributed in discrete particles having non-zero mass and consequently non-zero inertia. This conclusion has been reached earlier on the basis of more sophisticated arguments by Cooper, Kelleher and Lee.[6] The proper treatment of the effects of a plasma on an atomic system requires a self-consistent field approach, such as that of Cauble, Blaha and Davis[7] (who give references to earlier work).

A third model that has been frequently employed is based on the idea that the plasma microfield limits the number of bound states of atomic systems. As this picture forms the basis for our treatment, we defer discussion of it until the next section.

CURRENT APPROACHES

It is essential to recognize that the dichotomy between bound and free electrons is too simple, since electrons in a plasma can be semi-bound; an early extensive discussion of this problem was given by Gündel.[8] For example, when a hydrogen atom is perturbed by a nearby proton, the electron can be unbound in the one-center system, but bound in the two-center system (i.e. H_2^+). More generally, an electron can be bound in complexes containing 2,3,4,... ionic centers, which persist for times on order of the mean interionic distance speeds divided by the relative speed of the ions. Busquet[9] has discussed the migration of electrons as the complexes form and dissolve. Thus we see a rough similarity to the band structure of solids, or, more closely, of the liquid state. More[10] has treated the quasi-bound electrons quantitatively as shape resonances in the short-range potential given by the ion-sphere model.

It is useful to characterize current efforts on these problems as based on either the physical, or the chemical, picture.[11] In the former, the theory starts with electrons and nuclei interacting through the Coulomb potential, and then derives the properties of composite particles (i.e. atoms, ion and molecules), an extraordinarily difficult problem which can be solved only by a tour de force of quantum statistical mechanics. Solutions to this problem, which must, of course, involve approximations, have been obtained recently by Rogers,[12] using the technique of activity expansions and by Ebeling[13] and his associates in East Germany, using a Green's function approach. We cannot discuss this work further, but some results of Rogers' will be given below.

In the chemical picture, one begins by assuming the existence and properties of composite particles and treats the interactions with the plasma as perturbations, using the familiar ideas of atomic physics. This is a phenomenological approach, involving such familiar concepts as collision cross sections and plasma microfields, and so has considerable scope for physical intuition (and failures thereof). The basic idea is to construct an expression for the free-energy which is then minimized with respect to the level populations of the composite particles.

The approach that we have exploited is known as the occupation probability method, in which an occupation probability w_i is inserted into the Boltzmann law and the partition function. Thus we rewrite Eqs. (1) and (2) as

$$N_i = w_i \exp(-\varepsilon_i/kT)/Z(T) \tag{13}$$

and

$$Z(T) = \sum w_i \exp(-\varepsilon_i/kT) \quad . \tag{14}$$

We can interpret w_i as the probability of finding the atom in state i relative to similar ensembles of non-interacting particles or as the factor by which interactions with the plasma reduce the phase space available to the atomic state in question. If $w_i \to 0$ sufficiently rapidly as $i \to \infty$, the internal partition function $\tilde{Z}$ is finite. If $w_i \to 0$ smoothly, this approach avoids the discontinuities in $\tilde{Z}$ that exist if all states are assumed to exist up to some maximum number which depends on the plasma parameters, a point of importance in equation of state calculations.

The crucial question is then: How does one calculate the w_i? A number of workers[8,14,15] have used models accounting for Stark ionization arising from the static plasma microfield or from a nearby ion. Others [8,16] have considered the broadening of bound states into the continuum, either directly or by means of "upwards cascade," caused by the uncertainty principle and the very large collision rate. Our work[1] has extended and refined the treatment of both of these mechanisms; here we concentrate on the first as it turns out to be the more important for stellar envelopes.

The basic idea of the Stark ionization picture is that a bound state i can exist only if the field strength F is smaller than some critical value F_i^c, which depends on the state i. The probability that the bound state i exists is then

$$w_i = \int_0^{F_i^c} dF\, P(F) \quad , \tag{15}$$

where P(F) is the microfield distribution function. We are then faced with two questions: 1) How does one calculate F_i^c? and 2) What is the appropriate microfield distribution function?

Saddle-point Estimates of F_i^c. Unsöld[14] estimated F_i^c in two ways. In one of these models the orbital electron (in a hydrogenic ion) was assumed to move in the sum of the Coulomb potential arising from the nuclear charge Z_a and the potential $\underline{F} \cdot \underline{r}$, where $\underline{F}$ is the magnitude of the microfield assumed to be spatially uniform and independent of time. This composite potential has a saddle point, as illustrated in Fig. 1. The assumption is that no bound states exist above the saddle point. (Although we now know this to be false, this picture provides a convenient basis for an improved treatment.) For a state of ionization potential χ, the critical field is then easily shown to be

$$F^c(\chi) = \chi^2/4Z_a \quad \text{a.u.} \tag{16}$$

Using the nearest-neighbor microfield distribution, and taking the product of the probabilities w for each perturber species p, Unsöld obtained

$$w(\chi) = \exp[-(32\pi/3)\, a_0^3\, \chi^{-3}\, Z_a^{3/2} \sum_p N_p\, Z_p^{3/2}] \tag{17}$$

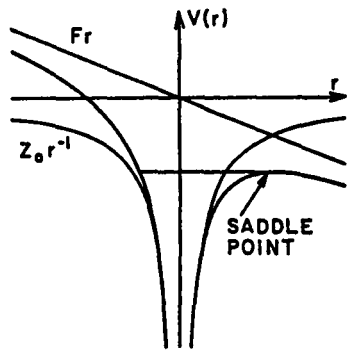

Fig. 1. Coulomb and uniform field potentials and their sum (heavy line), in the plane containing the saddlepoint.

For a hydrogenic ion we have

$$\chi_n = Z_a^2/2n^2 \text{ a.u.} \quad , \tag{18}$$

$$F_n^c = Z_a^3/16n^4 \tag{19}$$

and thus

$$w_n = \exp(-(n/n^*)^6) \quad , \tag{20}$$

where

$$n^* = [(256\pi/3)\, a_0^3\, Z_a^{-3/2} \sum_p N_p\, Z_p^{3/2}]^{-1/6} \quad . \tag{21}$$

Thus w_n is unity for state with $n \ll n^*$, and cuts off <u>very</u> sharply for $n \sim n^*$.

Unsöld's second model considered the saddle point between the overlapping Coulomb potential wells of two ions of charge Z_a and Z_p, separated by a distance ρ. By integrating the nearest-neighbor distribution for values of ρ larger than the critical separation which gives a saddlepoint at a specified value of χ, Unsöld obtains again Eq. (20), but with a somewhat larger value of n^*. We do not consider this model further, because of the difficulty mentioned above that an electron unbound in a one-center system can still be bound in the two-center system. Moreover, because this model accounts only for the nearest neighbor in a fundamental way, it drastically overestimates the weaker field.

<u>Additional Physical Mechanisms</u>. One quickly comes to realize that a great deal of physics is missing from the static saddle-point model. The simple model just discussed assumes that the component of angular momentum m along the field direction is zero. We have found

an exact solution to saddlepoint equations[17] for $m \neq 0$ which shows that F^c could be increased by as much as 2.7. Moreover, allowing for the Stark shift of the energy level changes F^c by a factor varying from 0.8 to 1.9, depending on the state in question. When the problem is examined in three dimensions,[18] it is readily seen that the electron can easily miss the saddle point, or is localized strongly on the "uphill" side of the potential surface, which would increase F^c substantially. A quantal saddle-point calculation[19] gives values of F^c differing from the classical values by substantial amounts, as does the use of accurate Stark ionization rates[20] which allows for tunneling. Finally, the intersection of adjacent Stark manifolds, which occurs for $n > 3$, is ignored; this mechanism turns out to be crucial.

Stark Level Interactions. Under plasma conditions, in which the microfield is spatially non-uniform, intersections of levels are of the "avoided crossing" type[21]; this is also caused by deviations of the ionic potential from pure Coulomb form,[21] including the relativistic interactions responsible for fine structure.[22] But because the microfield oscillates in both amplitude and direction with a wide spectrum of frequencies, Landau-Zener crossings are probable; electron collisions may increase this probability. The dynamical response of an atom to an oscillating microfield is sketched in Fig. 2, which shows only the extreme members of the Stark manifolds. As the field increases through zero, an electron with principal quantum number n in a "rising" state will traverse the intersection point if the field amplitude exceeds F^c_n, and has a non-zero chance of making a transition to a state of principal quantum number n+1. If it does not, it has further opportunities at subsequent sweeps through this point. If it does jump to $n + 1$, then the process occurs again in the opposite sweep of the field and a transition to $n + 2$ is possible. Thus if the field amplitude exceeds F^c_n, an atom in state n quickly ionizes, i.e. the state cannot exist.

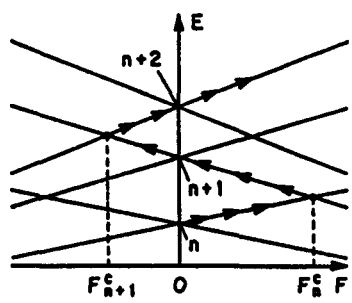

Fig. 2. Schematic Stark energy level diagram for states with principal quantum numbers n, n+1 and n+2; only the extreme states of each manifold are shown. F^c_n and F^c_{n+1} are the fields at which the nth and (n+1)-st manifold first intersect with the next higher. Arrows show the path followed by ionizing electrons.

In experimental studies of Stark ionization[23] of sodium in microwave fields, it was found that

$$F_n^c \sim \begin{cases} n^{-5} & m = 0,1 \\ n^{-4}, & m = 2 \end{cases} \quad . \tag{22}$$

However, in plasma the fluctuations of the microfield in <u>direction</u> mix m-states, so that "all go when the first goes," i.e. $F_n^c \sim n^{-5}$ should be valid for all m. This relation is in agreement with the field strength at the crossing of the outer members of the n-th with the (n+1)-st manifolds, which from first order perturbation theory is

$$F_n^c = (Z_a^3/3)\ (n+\tfrac{1}{2})/[n^2(n+1)^2(n^2+n+\tfrac{1}{2})] \underset{n\to\infty}{\sim} Z_a^3/3n^5 \quad . \tag{23}$$

For non-hydrogenic systems, states with moderate and large values of n are dominated by substates with large ℓ-values, for which the quantum defects are small. Thus the use of this theory for non-hydrogenic systems is at least reasonable.

The value of the critical field given in Eq. (23) is identical to that found by Inglis and Teller[24] for the merging of the spectral lines in a sequence. However, the physical processes are different, for Inglis and Teller were concerned with the field at which the states overlapped, which depends only on the amplitude of the microfield, while for state dissolution the <u>oscillations</u> of the microfield play a primary rule.

<u>Microfield distribution</u>. The choice of the microfield distribution function depends on two parameters: 1) The correlation among plasma particles, which is <u>unimportant</u> if

$$\Gamma \equiv \text{potential energy/kinetic energy} = Z_a Z_p\ e^2/r_0\ kT \ll 1 \ ; \tag{24}$$

2) The number of ionic species in the plasma. For stellar envelopes $\Gamma < 1$, so we use the Holtsmark distribution function and use an <u>ad hoc</u> rescaling procedure to allow for ions other than protons (since hydrogen is by far the dominant element in the normal, i.e. solar, abundance distribution). We distribute the positive charge uniformly over all ions. Thus our expression for w_n is

$$w_n = \int_0^{\beta_n^c} d\beta\ P_H(\beta) \quad , \tag{25}$$

where $P_H(\beta)$ is the Holtsmark distribution function,

$$\beta_n^c \equiv F_n^c/F_0 \tag{26}$$

and

$$F_0 = Z_p/r_0^2 + N_e N_{ion}^{-1/3}\ (4\pi a_0^3/3)^{2/3} \quad , \tag{27}$$

where the last expression allows for the rescaling just described.

Rational approximations are available[25] for the cumulative of $P_H(\beta)$, as well as for the function itself and its derivative. Inserting for F_n^c the estimates discussed above, and remembering that states with $n < 3$ rigorously do not cross, we have

$$\beta_c = K_n \, (x_n^2/4Z_a) \, (4\pi a_0^3/3)^{-2/3} \, N_e^{-1} \, N_{ion}^{1/3} \tag{28}$$

where

$$k_n = \begin{cases} 1 & , \; n \le 3 \\ (16/3)\,(n/n+1)^2\,(n+7/6)\,(n^2+n+1/2)^{-1} & , \; n > 3 \end{cases} \tag{29}$$

Figure 3 shows w_n for H and He^+ in plasmas with a abundance ratio He/H = 0.1, temperature T = 1.5 eV (17410 K) and $\rho = 10^{-4}$, 10^{-3}, 10^{-2} gm cm^{-3}.

Comparison with experiment. In view of the assumptions and approximations involved in obtaining our expression for w_n, it is of crucial importance to compare our results with experiment whenever possible. Unfortunately, relevant experimental data are very rare. However, we do have available a unique source of data from the accurate hydrogen line profiles measured by Wiese, Kelleher and Pacquette.[4] Hydrogen line profiles using the above theory have been

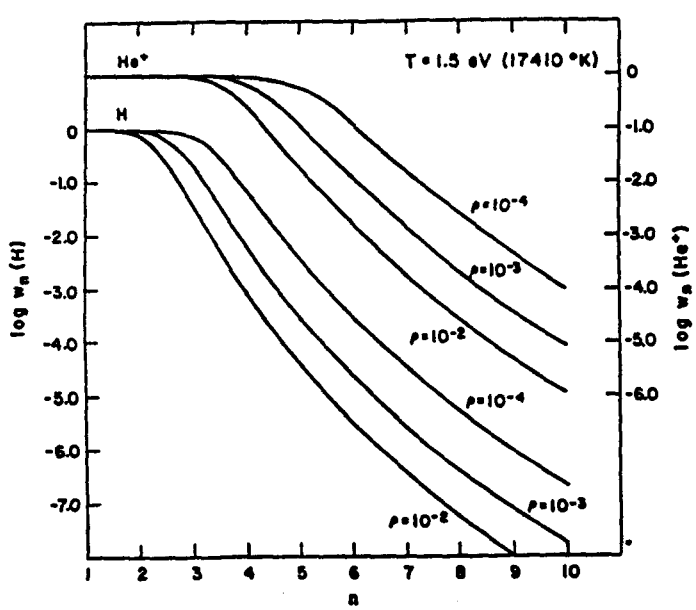

Fig. 3. Occupation probabilities w_n vs. n for H and He^+ in plasmas with T = 1.5 eV and densities = 10^{-4}, 10^{-3}, 10^{-2}. Corresponding values of the electron density are $N_e = 6.2 \times 10^{12}$, 2.2×10^{19} and 6.0×10^{19}, respectively.

calculated by Däppen, Anderson and Mihalas,[26] who also give prescriptions for accounting for the contribution of the "dissolved" states to the optical absortivity and emissivity. Their calculation involves no free parameters. The comparison with experiment is shown in Figs. 4 and 5, which correspond to $N_e = 1.8 \times 10^{16}$ cm^{-3}, $T = 1.0 \times 10^4$ and $N_e = 9.3 \times 10^{16}$, $T = 1.33 \times 10^4$ respectively. Calculations are presented for two choices of the occupation probabilities, those given by an earlier form of our theory, and an ad hoc choice, $w_n = 1$, $n \leq 30$, and vanishing otherwise. There is a substantial contribution to the continuum from electrons from the dissolved

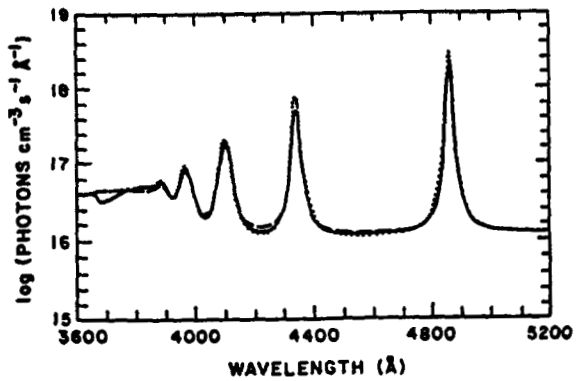

Fig. 4. Emissivities vs. wavelength for $N_e = 1.8 \times 10^{16}$ cm^{-3} and $T = 1.0 \times 10^4$ (after Däppen et al.[26]). Dotted line is experimental data of Wiese et al.,[4] long dashed line is occupation probability formalism, and solid line is ad hoc occupation probability described in text.

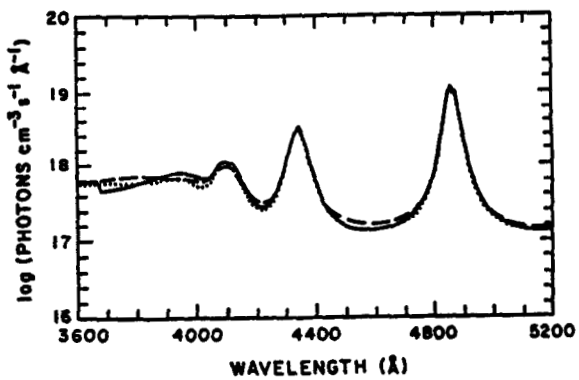

Fig. 5. As Figure 4, but for $N_e = 9.3 \times 10^{16}$ cm^{-3} and $T = 1.33 \times 10^4$ (after Däppen et al.[26]).

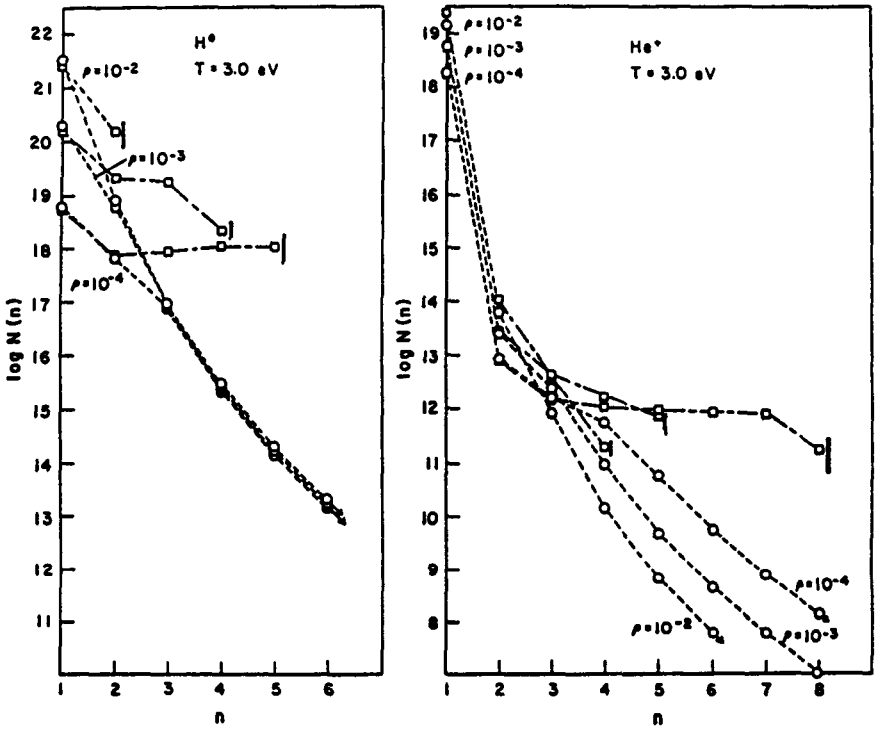

Fig. 6. Level populations vs. principal quantum number n for H and He^+, in plasmas with He/H = 0.1, T = 3.0 eV and $\rho = 10^{-4}$, 10^{-3}, 10^{-2}, corresponding to $N_e = 3.8 \times 10^{19}$, 2.4×10^{20} and 1.1×10^{21} cm^{-3}. Occupation probability values o---o, activity expansion □-- - --□.

states. The agreement between theory and experiment is, of course, not proof that the theory is correct, but at least there is no experimental evidence to date that it is seriously in error.

Comparison With Activity Expansion Results. Forrest Rogers has kindly supplied unpublished level populations for hydrogen-helium plasmas at various temperatures and densities, obtained from his activity expansion calculations.[12] Figure 6 shows the populations of H and He^+ as a function of principal quantum number n for H-H_e plasmas with He/H = 0.1 at a temperature of 3. eV, for densities of $\rho = 10^{-4}$, 10^{-3}, 10^{-2}, corresponding to electron densities $N_e = 3.8 \times 10^{19}$, 2.4×10^{20} and 1.1×10^{21} cm^{-3}, respectively. Full equation of state calculations were carried out using the activity expansion and occupation probability methods, so that the ionization equilibria in each case are internally consistent. The results are in fair agreement for the ground state, but differ quantitatively and qualitative-

ly for $n \geq 3$. In the activity expansion calculation, states for which no points are shown do not exist. Although it can not be seen from these figures, the (n,ℓ) populations from the activity expansion calculation show substantial deviations from the $2\ell+1$ dependence usually expected for atoms in thermal equilibrium.

These differences underline the urgent need for experimental guidance in this area. In particular, the direct measurement of level populations in low-lying states of atoms and ions in dense plasmas would be most valuable. Perhaps this could be done in laser absorption experiments, possibly using phase sensitive detection to overcome the plasma noise problems.

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