Semiclassical method for calculating the kinetics of radiative transitions in an atom

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An analytic method is developed for calculating the two-dimensional populations (the two dimensions are n and l) of highly excited atomic states. All possible radiative cascades are taken into account. The solutions reveal scaling laws for the populations and agree well with numerical quantum-mechanical calculations.

1. In several problems of practical importance, it has recently become necessary to carry out calculations on radiative cascades between highly excited (Rydberg) states (calculations of the populations of atomic levels excited by stepwise laser transitions, the luminous emittance of lines of multiply charged ions in a hot, low-density plasma, etc.; see, e.g., Ref. 1). Analytic calculation methods for radiative cascades have been developed only for the one-dimensional case[2,3] (in the principal quantum number n), in which the levels are assumed to be filled in a statistically weighted fashion in accordance with the orbital angular momentum l. For two-dimensional populations (in n and l), numerical quantum-mechanical calculations have been carried out[4] for the case of a photorecombination population source. As more levels are included in the cascade, these calculations becomes overly laborious and (as can be seen from Ref. 4) less accurate. The alternative use of the purely classical analytic description of a radiative cascade[5] is inadequate here (more on this below).

In this letter we develop a semiclassical method for calculating the populations $f(n,l)$ of atomic levels, taking all possible radiative cascades into account. The analytic solutions derived below make it possible to determine universal parameters of the problem and to thereby extract scaling laws for the populations, which are found to agree well with the numerical calculations.[4] The results derived below hold for a low-density plasma; one-dimensional populations (along n) of atomic levels were calculated in Ref. 6 for the case of a dense plasma, in which collisional processes are dominant.

2. We begin with the standard population balance equation[2–4]

$$\sum_{n'=n+1}^{\infty} \sum_{l'=l\pm 1} f(n', l') W(n', l' \to nl) - A(n, l) f(n, l) + q(n, l) = 0, \qquad (1)$$

where $W(n'l' \to n\,l)$ is the probability for the radiative transition $n'l' \to n\,l$, q is the external population source, and $A \equiv \Sigma_{n'=l+1}^{n-1} \Sigma_{l'=l\pm 1} W(nl \to n'l')$ is the total rate of the radiative decay of the level $n\,l$.

For $n \gg 1$ the summation over n in (1) can be replaced by an integration, and for $l \gg 1$ we can expand $f(n',l')$ near the point $l' = l$, with the ultimate result that (1) leads to an equation which is an integral equation in n and a differential equation in l. This integro-differential equation in turn "breaks up" into a one-dimensional integral equa-

tion (2) in the interval $x_m \equiv (l+1/2)^3/6n^2 \lesssim 1$ (which includes $x_m \ll 1$) and a two-dimensional differential equation (5) at $x_m \gg 1$; both of these equations are given below. At the boundary between these two regions, these solutions are joined unambiguously.

3. For $x_m \lesssim 1$ and $l \gg 1$, we can use the Kramers approximation ($l \ll n$) for W. In this case, the variable l appears as a parameter in an integral (over the energy) equation for $f(E,m)$ (since the rate of loss of angular momentum upon radiation is small in comparison with the rate of energy loss) [$G_0(x) = x(K^2_{1/3}(x) + K^2_{2/3}(x))$, where K_ν is the modified Hankel function]:

$$\int_0^{x_m} G_0(x)\, f\left(E\left(1-\frac{x}{x_m}\right), M\right) dx - f(E,M) \int_0^{\infty} G_0(x)\, dx = Q \equiv \pi q(n,l)/\sqrt{3} A(n,l), \tag{2}$$

where $E_{\text{a.u.}} = 1/2n^2 < 0, M = \hbar(l+1/2)$, $A_{\text{a.u.}} = 4[\sqrt{3}\,\pi n^3 c^3 (l+1/2)^2]^{-1}$. For small values of x_m, the integral term in (2), which describes the filling of a level by a cascade mechanism, is unimportant, and the population is determined exclusively by direct filling: $f = q/A$. Solving Eq. (2) by the Laplace method [$\bar{f}(s) = \bar{Q}(s)/sG_2(s)$, where s is the Laplace variable conjugate to x_m], and using the approximation

$$G_2(x) \equiv \int_x^{\infty} G_0(x')\, dx' = xK_{1/3}(x)K_{2/3}(x) \approx \alpha e^{-2x}, \quad \alpha = \frac{\pi^2}{6} \propto \frac{\pi}{\sqrt{3}} \approx 1,7, \tag{3}$$

which holds within 10%, we find

$$f_{nl} = q_{nl}/A_{nl} + \int_{n+1}^{\infty} q(n',l)/|\dot{n}(n',l)|\, dn', \quad \dot{n}_{\text{a.u.}} = -[c^3 (l+1/2)^5]^{-1}, \tag{4}$$

where the function $\dot{n}$ is related to the rate of radiative energy loss by a classical particle. It can be seen from (4) that where approximation (3) is applicable (as discussed in Section 6 below), the cascade filling of a level turns out to be purely classical.

4. At $x_m \gg 1$, Eq. (1) becomes the corresponding purely classical kinetic equation,[5] after an expansion of $f(n',l')$ around the point $n\,l$ within the integral:

$$\dot{E}\,\partial f/\partial E + \dot{M}\,\partial f/\partial M - \dot{M} f/M = q, \tag{5}$$

where $\dot{E}$ and $\dot{M}$ are the rates of the radiative loss of energy and angular momentum. In order to derive (5), it is necessary to retain, along with the classical limit of the function W, the first quantum correction to it (cf. Ref. 7), since the contributions from the leading terms cancel out [see, e.g., Eq. (2) at $x_m \gg 1$]. This classical case corresponds to a continuous flow of electrons through atomic levels, in which the role of phase trajectories is played by the characteristics of Eq. (5), specified by

$$\tau(E,M) \equiv M^{-3}(1 - 2EM^2/mZ^2e^4) \equiv M^{-3}\epsilon^2 = \text{const}, \tag{6}$$

where ϵ is the eccentricity of the orbit. By solving Eq. (5) by the method of characteristics we can extend the classical cascade filling of levels into the non-Kramers region ($l \sim n$) and derive a semiclassical solution of Eq. (1) for arbitrary l/n:

$$f_{nl} = q_{nl}/A_{nl} + M \int_{n+1}^{\infty} q(n', M(\tau,n'))/|(M(\tau,n')\,|\dot{n}(n', M(\tau,n'))|)\, dn', \tag{7}$$

where $M(\tau,n)$ is found from (6).

5. Applying the results of Sections 3 and 4 to the photo-recombination filling of levels from the continuum, with a Maxwellian distribution of free electrons, we finally find the result for the population $(b_n l)$ of level nl (in units of thermodynamic-equilibrium distribution):

$$b_{nl} = \frac{2}{2+x_T\epsilon^2} e^{-E/T} + \frac{1}{\alpha}\int_{x_m}^{\infty} e^{-yx_T} y(K_{1/3}^2(y) - K_{2/3}^2(y))\,dy, \tag{8}$$

$$x_T = \frac{3}{(TM^3)_{\text{a.u.}}}$$

[see (2), (3), and (6) for the notation]. Expression (8) is approximate ($\lesssim 10\%$) at $n, l \gg 1$ only because of our use of approximation (3) in the Green's function of Eq. (2). In the source itself, which in this case is of the same nature as the radiative cascade itself, approximation (3) was not used [the latter is responsible for the second term in (8)].

From (8) we find some scaling laws for the population. For $x_m \ll 1$ and $x_T \gg 1$, $f_{n\,l}$ depends on only the two variables x_m and x_T [only in this region is the second term in (8) important]. When x_m or $1/x_T$ reaches a value ~ 1, on the other hand, only the first term remains, and $f_{n\,l}$ depends on the one parameter $x_T\,\epsilon^2$.

A comparison of distribution (8) with the results of the numerical calculations[4] shows that the former also holds for small values $l \sim 1$. The validity of the scaling relation in terms of the variable x_T is confirmed by replotting the data of Ref. 4 with approximately equal values of x_m and ϵ as functions of x_T (Fig. 1). The difference between the resulting curve and that calculated from (8) does not exceed 20%. Table I compares the populations from Ref. 4 with those from (8) for $n = 6$ and $l = 1$–5 (the maximum discrepancy is 30% at $l = 1$).

Distribution (8) differs significantly from a statistically weighted distribution [b_{nl}

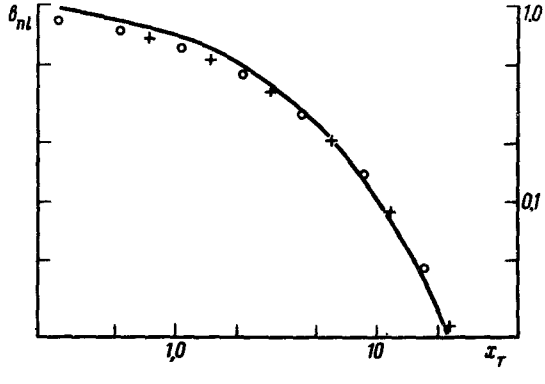

FIG. 1. Universal dependence of the level populations b_{nl} on the parameter x_T, according to (8). This dependence has been confirmed by numerical calculations[4] for the states $n = 10$, $l = 3$ (circles) and $n = 6$, $l = 2$ (crosses) ($x_m = 0.072$).

TABLE I. Populations of the levels $n = 6$, $l = 1$–5 for $T = 10^4$ K according to calculations from (8) and the calculations of Ref. 4.

b_{nl} \ l	1	2	3	4	5
(8)	0.060	0.20	0.38	0.53	0.62
[4]	0.0842	0.214	0.372	0.524	0.599

$= \phi(n)$], especially at low temperatures ($x_T \gg 1$). The population f_n calculated from (8), integrated over l, depends on the parameter Tn^3, not Tn^2 as in the one-dimensional case.[2]

6. This semiclassical method for calculating populations provides an algorithm for summing an infinite number of terms in a quantum cascade matrix and in the general quantum-mechanical case. The algorithm is based on the classical nature of the cascade filling of levels by multiquantum (multistep) transitions. The particular number of transition steps N for which the semiclassical description is valid depends on the particular source and the specific values of n and l. The accuracy of the final result is determined by the relative change in the result when one more term, corresponding to a filling of levels by $(N+1)$-step transitions, is distinguished in the classical cascade. For example, the distinction of single-quantum transitions gives us

$$f = q/A + \langle q \rangle / A + M \int \frac{\langle q \rangle \, dn'}{|\dot{n}| \dot{M}(\tau, n')}, \qquad (9)$$

$$\langle q \rangle = \sum_{n'=n+1}^{\infty} \sum_{l'=l\pm 1} q(n', l') \frac{W(n'l' \to nl)}{A(n', l')}.$$

Expression (9) is most suitable for levels near a selective source [$q \propto \delta(n - n_0)\delta(l - l_0)$]. For a photorecombination source, the results in (8) differ from those calculated from (9) by less than 10%.

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