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SEMICLASSICAL THEORY OF ION STOPPING⁽¹⁾

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Résumé - Le calcul du ralentissement d'ions multichargés nécessite l'élaboration de nouvelles techniques pour la détermination des forces d'oscillateur. Nous décrivons, ici, une méthode semi-classique donnant les éléments de matrice électroniques et les forces d'oscillateur correspondantes. La méthode est évaluée par comparaison avec des résultats connus pour le potentiel de Coulomb. Elle est numériquement robuste et raisonnablement précise pour toutes les transitions considérées. Elle est appliquée par le potentiel de Bethe pour l'excitation de l'hydrogène.

Abstract - There is a need for a convenient method to calculate oscillator strengths for highly charged ions for application to the calculation of stopping powers. In this paper we describe a new semiclassical method for finding electron matrix-elements and oscillator strengths. The method is tested by comparison with known results for the Coulomb potential and proves to be numerically robust and reasonably accurate for all transitions examined. It is applied to calculate the hydrogen Bethe excitation potential.

1 - STOPPING-POWER AND OSCILLATOR-STRENGTHS

We begin with a brief discussion of the connection between radiative opacity and fast-ion stopping powers.¹

The electromagnetic field of a fast ion is approximately equivalent to a cluster of virtual photons carried along by the ion. When the ion penetrates a plasma target, some of the virtual photons will be absorbed by electrons of the target material. After being absorbed, the virtual photons are regenerated by the electric current of the fast ion, but the energy required for this is taken from the center-of-mass motion of the ion and therefore the absorption of virtual photons is equivalent to electronic energy-loss. From this point of view, dE/dx can be regarded as an average or mean opacity for absorption of virtual photons.

The quantitative expression of this intuitive picture is obtained by calculating the absorption of virtual photons. We use the frequency spectrum of virtual photons,

$$N(h\nu) = \frac{2\alpha}{\pi h\nu} Z_1^2 \left(\frac{\xi}{v}\right)^2 \log \left(\frac{a m v^2}{h\nu}\right) \quad (1)$$

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Here Z_1 is the ion charge, v is its velocity, c is the speed of light, $\alpha = e^2/\hbar c$ is the fine-structure constant and the coefficient a is a dimensionless number of order unity.² We employ a minimum impact parameter $b_{\min} = \hbar/mv$, as is normal for high-velocity ion interactions.

The cross section per gram for line transitions of bound electrons of the target plasma is determined by the bound-bound absorption opacity, which we can write in the notation of the average-atom model,

$$\kappa_{\nu}^{bb} = \frac{\pi e^2 \hbar}{mc A m_p} \sum_{n, n'} P_n \left(1 - \frac{P_{n'}}{D_{n'}}\right) f_{nn'} I(\hbar\nu) \quad (2)$$

There are also bound-free and free-free absorption processes. P_n is the number of electrons in the lower shell of principal quantum number n and $(1 - P_{n'}/D_{n'})$ is a correction for partial filling of states of the final shell n' ($D_{n'} = 2n'^2$). $I(\hbar\nu)$ is the line absorption profile, which can be replaced by a delta-function, $I(\hbar\nu) \approx \delta(\hbar\nu - (E_{n'} - E_n))$, and $f_{n, n'}$ is the absorption oscillator strength, summed over states in the final shell n' and averaged over states of the initial shell n .

The energy lost by absorption of virtual photons is then given by¹

$$-\frac{dE}{dx} = \int \hbar\nu N(\hbar\nu) \rho \kappa_{\nu} d\hbar\nu \quad (3)$$

This integral gives the following expression for the energy-loss:

$$-\left(\frac{dE}{dx}\right)_{bb} = \frac{4\pi Z_1^2 e^4}{mv^2} n_i \sum_{nn'} f_{nn'} P_n \left(1 - \frac{P_{n'}}{D_{n'}}\right) \log\left(\frac{amv^2}{\hbar\nu}\right) \quad (4)$$

This result is the bound-bound part of the Bethe formula for high-velocity energy-loss.²

Several interesting physical questions arise at this point: Does it matter that the photons considered are virtual photons rather than real photons? Should this slightly modify the energy-conservation relation? When we apply these equations to plasmas, should we include the downward transitions, in which an excited atom apparently gives energy to the fast ion? Could there be stimulated emission by excited target ions caused by virtual photons of the fast ion? While we are inclined to answer these questions in the affirmative, perhaps further study is required.

Usually one writes the stopping power due to bound electrons as

$$-\left(\frac{dE}{dx}\right)_b = \frac{4\pi Z_1^2 e^4}{mv^2} n_i N_B \log\left(\frac{amv^2}{\bar{I}}\right) \quad (5)$$

where N_B is the number of bound electrons per atom (ion) and \bar{I} is the mean excitation-ionization potential, defined by:

$$\log \bar{I} = \frac{\sum_{nn'} f_{nn'} P_n (1 - P_{n'}/D_{n'}) \log(E_{n'} - E_n) + \text{bf term}}{\sum_{nn'} f_{nn'} P_n (1 - P_{n'}/D_{n'}) + \text{bf term}} \quad (6)$$

At this point the photoelectric (bound-free) contribution is merely indicated in a schematic fashion. Because of the f-sum rule, the denominator in Eq. (6) is simply $N_B = Z - Q$, the number of bound electrons.

The evaluation of \bar{I} requires the energy-levels and absorption oscillator-strengths for transitions between these eigenstates.³

There is a simple approximation for hydrogen-like ions, given by Bethe and Salpeter⁴ and Menzel and Pekeris⁵:

$$f_{nn'} = \frac{32}{3\pi\sqrt{3}} \frac{1}{n^5} \frac{1}{n'^3} \frac{1}{(1/n^2 - 1/n'^2)^3} \quad (7)$$

Equation (7) is accurate to within a factor of two, for example, the $1s \rightarrow 2p$ matrix-element is ($a_0 = \hbar^2/me^2 = \text{Bohr radius}$)

$$R_{1s}^{2p} = \int \Psi_{1s} r \Psi_{2p} dr = \frac{1}{\sqrt{6}} \left(\frac{4}{3}\right)^4 \frac{a_0}{Z} = 1.2903 \frac{a_0}{Z} \quad (8)$$

which gives an exact oscillator-strength of $f(1s-2p) = 4 (2/3)^9 = 0.4162$, while Eq. (7) gives $f(1,2) = 0.5808$, which is 40% high. Equation (7) is more accurate for states with higher quantum numbers.

Equation (7) can be derived by extrapolation of the Kramers' absorption cross-section for Bremsstrahlung to negative energies of both initial and final states for the absorbing electron.^{1,6}

Reference (1) also generalizes Eq. (7) to non-hydrogenic atoms, within the context of the screened hydrogenic model. The electron energy-levels are taken to be

$$E_n = - \frac{Q_n^2 e^2}{2a_0 n^2} + E_n^{(0)} \quad (9)$$

where Q_n is the effective charge for electrons of shell n

$$Q_n = Z - \sum_m \sigma_{nm} P_m \quad (10)$$

$E_n^{(0)}$ is an outer-screening correction (also a simple function of P_n, Q_n). Repeating the extrapolation to negative energies with this representation of the energies, one finds:

$$f_{nn'} = \frac{4}{3\pi\sqrt{3}} \frac{Q_n^4 Q_{n'}^2}{n^5 n'^3} \left(\frac{e^2/a_0}{E_{n'} - E_n} \right)^3 \quad (11)$$

For the hydrogenic case $E_n^{(0)}$ is zero and the effective charges Q_n are equal to Z ; in this case the Q_n 's cancel and Eq. (11) reduces to Eq. (7). For $1s-np$ transitions of partially ionized heavy atoms, Eq. (11) can differ from Eq. (7) by large factors (5 to 20) and is systematically more accurate.¹ However Eq. (11) still differs from the best theoretical calculations by factors of two due to quantum and relativistic effects.

For energy-loss calculation, Eq. (11) is not sufficient because $n\ell \rightarrow n'\ell'$ transitions are important and subshell splitting greatly affects the answer. One would like the detailed

oscillator-strength which describes transitions between states of specified angular momentum, $f(nl, n'l')$.

The oscillator-strength is required for most calculations of atomic processes in hot plasmas: calculation of stopping-power, calculation of emission and absorption of radiation and in many approximate formulas for electron-impact excitation and ionization. For this reason we have studied the possibility of developing a formula more detailed and accurate than Eq. (11) which would retain its simplicity and generality.

Although this problem has attracted a great deal of attention,⁷⁻¹⁸ the existing results are not very satisfactory.

There are interesting semiclassical results based on Fourier analysis of the classical motion on an average trajectory corresponding to average quantum numbers n_c, l_c of the electron.¹¹⁻¹³ These methods assume $|n - n'| = \Delta n \ll n$. For the hydrogenic system the result is an expression involving Bessel functions related to Kramers' original result; with an optimal choice of n_c this method is very accurate. Unfortunately the results apply to the Coulomb potential, and the generalization is not simple.

We are interested in two types of non-Coulomb potentials: those with atomic screening by core electrons, and those with external plasma screening by free electrons and ions of a dense plasma environment.

For these problems we have studied the use of WKB methods. However the available WKB methods (e.g., reference 14) fail for simple problems such as the Coulomb potential or the linear harmonic oscillator. Something better is needed.

2 - WKB APPROXIMATION FOR OSCILLATOR STRENGTHS

It is well-known that the semiclassical WKB approximation gives very good (exact) results for the energy-levels of a nonrelativistic hydrogenic ion. We will extend the WKB method to calculate matrix elements. Our results are within a few percent of the exact answers for transitions in the Coulomb potential, even for low quantum numbers such as the $1s \rightarrow 2p$ transition.

The method developed here applies to other matrix-elements and other potentials without modification. It has clear advantages for transitions to/from states with high quantum numbers or for sums over many transitions. Because the method is numerically robust we believe it will succeed for many other applications.

In the WKB theory for spherically-symmetric systems, the radial wave-function Ψ_{nl} is a sum of incoming and outgoing waves,

$$\Psi_{nl}(r) = \frac{1}{2} \left[\Psi_{nl}^{(-)} + \Psi_{nl}^{(+)} \right] \quad (12)$$

$$\psi_{n\ell}^{(\pm)}(r) = \frac{c_{n\ell}}{\sqrt{q(r)}} \exp(\pm i[\varphi(r) - \pi/4]) \quad (13)$$

$$\varphi(r) = \int_{r_1}^r q(r') dr' \quad (14)$$

$$q(r) = \sqrt{\frac{2m}{\hbar^2} \left(E_{n\ell} + eV(r) - \frac{\hbar^2}{2m} \frac{\left(\ell + \frac{1}{2}\right)^2}{r^2} \right)^{1/2}} \quad (15)$$

where $E_{n\ell}$ is the energy eigenvalue, $V(r)$ is the electrostatic potential, and $r_1 = r_1(n, \ell)$ is an inner turning-point, defined by $q_{n\ell}(r_1) = 0$. The quantity $q_{n\ell}(r)$ is the wave-vector, proportional to the radial velocity, and φ is the phase-integral, related to the action function of classical mechanics. In our work we allow the radius r to be a complex variable.

The normalization $c_{n\ell}$ of the wave-function can be determined by the WKB normalization rule

$$|c_{n\ell}|^2 = \frac{2}{\int_{r_1}^{r_2} \frac{dr}{q(r)}} = \frac{2m}{\pi \hbar^2} \frac{dE_{n\ell}}{dn} \quad (16)$$

To calculate the matrix-element of r with WKB wave-functions, we first write

$$R_{n'\ell'}^{n\ell} = \frac{1}{2} \operatorname{Re} \int_0^\infty [\psi_{n\ell}^{(+)}(r) r \psi_{n'\ell'}^{(-)}(r)] dr \quad (17)$$

This is a good approximation for large n, n' because the omitted terms, integrals of $\psi_{n\ell}^{(-)}$, $\psi_{n'\ell'}^{(-)}$, and $\psi_{n\ell}^{(+)} \psi_{n'\ell'}^{(+)}$, oscillate rapidly and symmetrically about zero. Physically, this can be understood as follows: the integral we keep,

$$\int [\psi_{n\ell}^{(-)}]^* r \psi_{n'\ell'}^{(-)} dr = \int \psi_{n\ell}^{(+)} r \psi_{n'\ell'}^{(-)} dr \quad (18)$$

corresponds to the transition of an incoming $n'\ell'$ electron onto an incoming $n\ell$ orbit. This is the transition involving the smaller change of momentum for the electron. The transition to an outgoing $n\ell$ orbit, corresponding to the integral

$$\int [\psi_{n\ell}^{(+)}]^* r \psi_{n'\ell'}^{(-)} dr = \int \psi_{n\ell}^{(-)} r \psi_{n'\ell'}^{(-)} dr \quad (19)$$

has a much smaller probability because it requires a reversal of the electron velocity. The photon momentum is negligible compared to the electron momenta, and so this reversal is extremely unlikely. (The probability is not exactly zero because momentum is not conserved in the electrostatic field of the nucleus.)

We use Eq. (17) for all values of n, n' and this is the core of our method.

The integrand in Eq. (17) has a saddle point at a complex radius r_{sad} defined by

$$G(r_{\text{sad}}) = 0 \quad (20)$$

where $G(r) = G(r; n\ell; n'\ell')$ is defined by

$$\begin{aligned}
 G(r) &= -i \frac{d}{dr} \log[\Psi_{n\ell}^{(+)}(r) r \Psi_{n'\ell'}^{(-)}(r)] \\
 &= q_{n\ell}(r) - q_{n'\ell'}(r) - \frac{i}{r} + \frac{i}{2} \left[\frac{q'_{n\ell}(r)}{q_{n\ell}} + \frac{q'_{n'\ell'}(r)}{q_{n'\ell'}} \right]
 \end{aligned} \quad (21)$$

For the Coulomb potential the saddle point can easily be found by a search (using Newton's rule) starting at

$$r_0 = \frac{4(\ell+1) a_0/Z}{\left[\frac{(\ell + \frac{3}{2})^2}{n^2} - \frac{(\ell + \frac{1}{2})^2}{(n')^2} \right]} e^{-i(0.9)} \quad (22)$$

In many cases Eq. (22) is an accurate approximation to the saddle-point, and this is not accidental, because r_0 is determined by the condition that the initial and final orbits should intersect with second-order contact. In any case, Eq. (22) is a reasonably good first guess of the solution of Eq. (20).

We note that the quantities $G(r)$, $G'(r)$ and r_{sad} are calculated directly from the potential $V(r)$, which must be known for complex values of radius r .

The saddle-point integration gives

$$R_{n\ell}^{n'\ell'} = \frac{1}{2} \text{Re} \left[\Psi_{n\ell}^{(+)}(r_s) r_s \Psi_{n'\ell'}^{(-)}(r_s) \sqrt{\frac{2\pi i}{G'(r_s)}} \right] \quad (23)$$

Evaluation of this expression requires integrating Eq. (14) to find the phase φ at the saddle-point. However the integration is not delicate because the integrand does not oscillate; even coarse zoning produces reasonable results.

The oscillator strength is now

$$f_{n\ell}^{n'\ell'} = \frac{2m}{\hbar^2} (E_{n'\ell'} - E_{n\ell}) \left[\frac{\max(\ell, \ell')}{3(2\ell+1)} \right] |R_{n\ell}^{n'\ell'}|^2 \quad (24)$$

for dipole-allowed transitions $n\ell \rightarrow n'\ell' \pm 1$. The factor in brackets arises from a sum over final states and average over initial states of given n, ℓ , n', ℓ' .

In the next section we will give examples, tests and applications of these formulas. Aside from simplicity and intuitive appeal, the main advantage of Eq. (23) is the possibility of extending it to non-hydrogenic systems where we obtain comparable results without a significant increase in the complexity of calculation.

3 - RESULTS AND APPLICATIONS

First we illustrate the success of Eq. (23) in calculation of dipole matrix-elements for the hydrogen atom.

For the $1s \rightarrow 2p$ transition, the WKB integral has a saddle-point at $r_s = (1.576 - 1.649i)a_0/Z$ where $a_0 = .529 \times 10^{-8}$ cm is the Bohr radius. Equation (23) gives

$$R_{1s}^{2p} = 1.251 (a_0/Z)$$

$$f_{1s}^{2p} = 0.3913$$

The nonrelativistic exact values are $R = 1.2903 a_0/Z$, $f = 0.4162$ while Kramers' formula gives $f = 0.5808$.

For matrix-elements of the series $1s \rightarrow np$ the error grows but does not exceed 10%. As will be seen from Table 1, other allowed dipole transitions have comparable accuracy. Extended comparison for a large variety of transitions shows that the method works very well.

TABLE 1

Transition	WKB $R(n, \ell, n', \ell')$	Exact R	percentage difference
$1s - 2p$	1.2510	1.2903	3.04
$1s - 3p$	0.4782	0.5167	7.45
$1s - 4p$	0.2796	0.3046	8.21
$1s - 5p$	0.1910	0.2087	8.49
$2s - 3p$	3.1439	3.0648	2.58
$2p - 3d$	4.6123	4.7480	2.86
$2p - 3s$	1.0019	0.9384	6.76
$3s - 4p$	5.7413	5.4693	4.97
$3p - 4d$	7.6298	7.5654	0.85
$3p - 4s$	2.6982	2.4435	10.42
$3d - 4f$	9.9253	10.2303	2.98
$3d - 4p$	1.4379	1.3023	10.41

The WKB method described here applies also to photoelectric absorption cross-sections. For transitions $1s \rightarrow \epsilon p$ to the continuum state with energy $\epsilon > 0$, angular momentum $\ell = 1$, we find a WKB cross-section which is consistently 15% lower than the exact nonrelativistic quantum cross-section for all energies from the threshold at 13.6 eV to several hundred eV, i.e., over a range of about 10^6 in cross-section. For transitions from higher bound-states the agreement is better.

Using the WKB results for line and photoelectric absorption, we have evaluated the Bethe logarithm for stopping of fast charged particles. Our result and the corresponding comparison values are:

$$\bar{I} \text{ (WKB)} = 14.5 \text{ eV}$$

$$\bar{I} \text{ (Exact)} = 14.8 \text{ eV}$$

$$\bar{I} \text{ (Kramers)} = 14.2 \text{ eV}$$

In evaluating this sum, we corrected the WKB and Kramers' values for their failure to exactly obey the f -sum rule. This correction is made by evaluating the sum of all oscillator strengths for transitions from the initial $1s$ state and then dividing by this to renormalize the oscillator strengths as in Eq. (6). Because the error in the WKB f -number is essentially a constant factor 0.85, at least after the first few transitions, the result of this correction is a rather accurate stopping-number.

A key application of our method will be to try to develop a rigorous fundamental justification for the well-known local-density model for stopping powers of fast charged particles. We have found that the matrix-element responsible for either bound-bound or photoelectric transitions reduces to a product of wave-functions evaluated at a single point, the saddle point. The sum over many transitions can then be transformed into an integral over saddle-points, and this integral is directly comparable with that assumed in the local-density theory.

We also applied our method to calculate quadrupole matrix-elements in the Coulomb potential, and find good agreement with the results of Oumarou et al.¹⁹

Does the method apply to non-Coulomb potentials, as in heavy atoms or ions? We have made a detailed study of Molybdenum ions in all charge states and find that the WKB method succeeds in reproducing the large changes in oscillator-strengths produced by bound-electron screening.

There have been a number of studies of the effect of plasma screening on the oscillator strengths for bound-bound and bound-free transitions.^{20,21} It is generally found that the strengths of individual transitions $n, \ell \rightarrow n', \ell'$ are strongly reduced as the state n', ℓ' comes close to the (lowered) continuum.

For the Debye-screened Coulomb potential,

$$V(r) = (Ze/r)\exp(-r/D) \quad (25)$$

Hohne and Zimmerman give some representative calculations. We compare with one of their cases:

	$D=20 a_0$	$D=\infty$	ratio	
$f_{1s \rightarrow 4p}^{WKB}$.0125	.0244	1.95	(26)
$f_{1s \rightarrow 4p}^{HZ}$.0145	.0290	2.00	

The agreement is again very impressive, considering the simplicity and convenience of the WKB calculation. In this case, while the oscillator strength $f(1s \rightarrow 4p)$ is reduced by a factor of two the saddle point is only changed a few percent by plasma screening. The main plasma effect is on the normalization of the WKB wave-function; in the screened Coulomb potential the $4p$ state relaxes outward and has reduced overlap with the inner $1s$ wave-function.

CONCLUSIONS

In this paper we have simply presented results of calculations based on Eq. (23). However the success of these calculations points to a surprising and important fact about semiclassical quantum mechanics. Results from Eq. (23) are better than those which are obtained if we normalize the wave-functions and compute the integrals directly according to the quantum-mechanical rules. That is to say, Eqs. (16) and (17) differ somewhat from the normal quantum rules, through the omission of terms like that in Eq. (19), and also work better.

We do not advocate changing the very successful rules of quantum mechanics, of course, but rather interpret the result as indicating that the omitted terms contain the most inaccurate portion of the WKB approximation, and physically correspond to processes which have very low probabilities.

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