

Exact expression of the impact broadening operator for hydrogen Stark broadening

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ABSTRACT

Aims. Recent measurements on the Stark broadening of radio recombination lines show values and trends in disagreement with conventional theories. Different attempts to explain those disagreements have not been successful for any of the employed theoretical models. In particular, the impact model that describes well the physical conditions at which the studied broadenings occur, shows a functional trend upon the principal quantum number of the studied transitions that does not correspond to the experimental observations.

Methods. High values of the principal quantum number require computable formulas for the calculation of transition probabilities. Some of those expressions have been published, leading to approximate formulas on the dependence of the line width versus the principal quantum number of the upper level of the transition.

Results. In this work an exact expression for the hydrogen Stark width in the frame of impact approximation is given.

Key words. plasmas – line: profiles

1. Introduction

Recent measurements of the width of radiofrequency spectral lines (Bell et al. 2000), together with discrepancies between experimental data (Smirnov et al. 1984; Smirnov 1985; Wilson & Jäger 1987; Sorochenko 1989; Bell et al. 2000) and conventional models (Baranger 1958a–c; Griem et al. 1959; Griem 1967, 1974) have stimulated the interest in this type of transitions. In this framework a controversy (see Griem 2005 and works cited therein) in explaining these disagreements without changing the usual frame of Stark broadening theories occur. To our knowledge, these discrepancies remain and have even been qualified as “mysterious” (Griem 2005; Watson 2006)¹.

In this context, approximate expressions are required to calculate transition probabilities between states with very high principal quantum numbers ($n > 100$) (Watson 2006; Hey 2006). This need rises because, although the analytical expressions of those transitions probabilities are known (Gordon 1929; Bethe & Salpeter 1957), they are not computationally manageable due to the large value of the factorials involved. In recent works (Watson 2006; Hey 2006) recurrence relations have been used to obtain transition probabilities that are much easier to evaluate. The investigation can be carried out in this way for high values of n ; although, as recognised by one of the authors (Watson 2006), the mystery of the discrepancy with the experiments cannot be solved with those approximate expressions.

In this work, the study relies on exact expressions of the width operator, resulting from the usual impact approximation frame, that will be given.

2. Initial expressions

Following Griem (Griem et al. 1959), the electronic collision contribution to the width of a transition $n \rightarrow n'$ is given by the expression

$$\Delta\omega_e = \pi\bar{v}_e N_e (\rho_{\min}^e)^2 + \frac{4\pi}{3\bar{v}_e} \left(\frac{\hbar}{m_e}\right)^2 N_e \frac{F_{nn'}}{S_{nn'}} \log\left(\frac{\rho_{\max}^e}{\rho_{\min}^e}\right), \quad (1)$$

where $\bar{v}_e = \sqrt{\pi kT/2m_e}$; N_e is the electron density; ρ_{\max}^e and ρ_{\min}^e are the upper and lower cutoff values respectively;

$$F_{nn'} = 2 \sum_{lm'l'm'\alpha} \left\{ \langle nlm|R_\alpha|n'l'm'\rangle \langle n'l'm'|R^2|n'l'm'\rangle \langle n'l'm'|R_\alpha|nlm\rangle \right. \\ \left. + \langle nlm|R_\alpha|n'l'm'\rangle \langle n'l'm'|R_\alpha|nlm\rangle \langle nlm|R^2|nlm\rangle \right. \\ \left. - 2 \sum_{l''m''l''m''\beta} \langle nlm|R_\alpha|n'l'm'\rangle \langle n'l'm'|R_\beta|n'l''m''\rangle \right. \\ \left. \times \langle n'l''m''|R_\alpha|n'l''m''\rangle \langle n'l''m''|R_\beta|nlm\rangle \right\}; \quad (2)$$

and

$$S_{nn'} = 2 \sum_{lm'l'm'\alpha} \langle nlm|R_\alpha|n'l'm'\rangle \langle n'l'm'|R_\alpha|nlm\rangle. \quad (3)$$

In these expressions, R_α , $\alpha = X, Y, Z$, is the position operator for the atomic electron. Parameters ρ_{\max}^e and ρ_{\min}^e are necessary to overcome the divergences occurring in the formal treatment due to the long and short range distances properties of the emitter-perturber interaction potential. The first term in (1) corresponds

¹ About this “mystery” see the comment in the conclusions of Griem (2005) that appears as a private communication with M. Bell.

to the so-called *strong collisions*, that give rise to a line broadening more similar to the Lorentz treatment of total coherence rupture. The second term fits the so-called *impact approximation*. It is obtained from the values of the dipole transition probabilities for transitions between the states of the emitter that, in our case, is a hydrogen atom.

The study of recombination transitions requires the evaluation of matrix elements of operator \mathbf{R} between states with high principal quantum number n . This is impossible when $n \gg 50$ (see Hey 2006 for a numerical solution of this problem).

Expression (1) is the quantum version of the classical expression labeled as (4) in Minaeva et al. (1968). These authors pointed out the difficulty of obtaining an analytical expression for the line widths. This is what we want to do here. We aim to obtain an exact and compact expression corresponding to the second term in (1), that is, corresponding to the Stark width due to the electron collisions that fulfil the requirements of impact approximation (rapid though weak collisions). The part corresponding to the first term in (1), the strong collisions term, will not be studied here.

In the next section, a review of the physical model as well as of the mathematical approximations leading to the so-called *impact model* in Stark broadening will be given. The expression used is not exactly (1), although it fits the same physical model. In particular, we do not analyze the final expression concerning the statistics of collisions, because it is removed from our subject: the dependence between the line widths and the principal quantum numbers of the levels involved in the transitions.

3. Review of impact model

Since Anderson's first works (Anderson 1949), dipole spontaneous-emission spectral profiles have been obtained from the autocorrelation function $C(t)$ of the dipole moment of the emitter $\mathbf{D}(t)$, via a Fourier transform of an average $\{ \}$ of the autocorrelation function over a given statistical sample:

$$I(\Delta\omega) = \text{Re} \frac{1}{\pi} \int_0^\infty dt \{C(t)\} e^{i\Delta\omega t}, \quad (4)$$

$$C(t) = \text{tr} [\mathbf{D}(t) \cdot \mathbf{D}(0)\rho] \quad (5)$$

$$\mathbf{D}(t) = U^+(t)\mathbf{D}(0)U(t), \quad (6)$$

where $U(t)$ is the time-evolution operator of the emitter, which satisfies the Schrödinger equation

$$i\hbar \frac{d}{dt} U(t) = H(t)U(t) = (H_0 + q\mathbf{E}(t) \cdot \mathbf{R})U(t), \quad (7)$$

where the Hamiltonian $H(t)$ includes the structure of the unperturbed states, H_0 , and the action of the charged perturbers via the dipole interaction, $q\mathbf{E}(t) \cdot \mathbf{R}$. In (7), $\mathbf{E}(t)$ is the electric microfield temporal sequence undergone by the emitter. The average indicated with $\{ \}$ in (4) is calculated over a representative sample of the microfield sequence. The trace in Eq. (5) averages over all initial states of the emitter. The density matrix ρ in (4) takes account of the population of the relevant states.

Following Baranger (1958a), the model considers the evolution of perturbers and emitter separately. In this sense, the plasma is considered as a "thermal bath" which alters the process of emission. The spectrum is then obtained as an incoherent superposition, i.e., a sum of intensities of the emission of independent atoms or ions.

In the majority of cases we study the transition between a group of upper and other of lower energy levels, and the relevant energy differences are in the optic or ultraviolet range. Since

in the astrophysical applications of interest, the energy transfers in collisions between emitter and perturbers are usually much smaller than this energy difference, we can consider that the collisions never induce transitions between the two groups of levels within the characteristic time of the emission (*no quenching approximation*). This is a limitation of the model, but, in spite of that, it has a very general scope, because we are dealing with the dominant effect for nearly all applications. Thus, we consider an evolution operator of the following form

$$U(t) = \begin{pmatrix} U_u(t) & 0 \\ 0 & U_l(t) \end{pmatrix} \quad (8)$$

where $U_u(t)$ and $U_l(t)$ are two evolution matrices which give the independent time-evolution of the states in the upper and lower groups respectively. The ρ matrix, which accounts for the populations at the beginning of the emission process, has the same structure: there are no coherences in the matrix. In the cases we will consider below, the states belonging to each group – upper and lower – have, in practice, equal populations and we can do without the density matrix.

On the other hand, when we consider the emission spectrum we are only interested in a rather small zone around the center of the spectral line. The frequency displacements are small enough to ignore dipole transitions that involve states not belonging to our two groups of states. Dipole transitions between two states of the same group, with energy differences much smaller than those of the optic transitions, can also be ignored. Accordingly, the dipole moment matrix in (6) is of the form

$$\mathbf{D} = \begin{pmatrix} 0 & \mathbf{d} \\ \mathbf{d}^+ & 0 \end{pmatrix}. \quad (9)$$

Hence, the trace in (5) can be written as follows

$$C(t) = \text{tr} (U_u^+ \mathbf{d} U_l \cdot \mathbf{d}^+) + \text{tr} (U_l^+ \mathbf{d}^+ U_u \cdot \mathbf{d}). \quad (10)$$

Matrices U_u and U_l are, respectively, the solution of equations

$$i\hbar \frac{d}{dt} U_u = H_u U_u, \quad i\hbar \frac{d}{dt} U_l = H_l U_l \quad (11)$$

with

$$\begin{aligned} H_u &= \mathcal{E}_u + H_{0u} + \mathbf{E}(t) \cdot \mathbf{R}_u \\ H_l &= \mathcal{E}_l + H_{0l} + \mathbf{E}(t) \cdot \mathbf{R}_l \end{aligned} \quad (12)$$

where $\mathcal{E}_u + H_{0u}$ and $\mathcal{E}_l + H_{0l}$ are the projections over the upper and lower subspaces, respectively, of the total unperturbed Hamiltonian for the emitter, and an equivalent notation is used for the projections of the operator \mathbf{R} over the same subspaces. Matrices \mathcal{E}_u and \mathcal{E}_l , both proportional to unity, take account of the energy gap between the two groups of states. We can proceed without these constants replacing U_u and U_l with $U_u \exp(-i\mathcal{E}_u t/\hbar)$ and $U_l \exp(-i\mathcal{E}_l t/\hbar)$ in (10) and, analogously, removing \mathcal{E} from H_u and H_l . We can then write the autocorrelation function as follows

$$\begin{aligned} C(t) &= e^{i\omega_0 t} \text{tr} (U_u^+ \mathbf{d} U_l \cdot \mathbf{d}^+) \\ &\quad + e^{-i\omega_0 t} \text{tr} (U_l^+ \mathbf{d}^+ U_u \cdot \mathbf{d}), \end{aligned} \quad (13)$$

with $\omega_0 \equiv (\mathcal{E}_u - \mathcal{E}_l)/\hbar$. Let us write

$$\text{tr} (U_l^+(t) \mathbf{d}^+ U_u(t) \cdot \mathbf{d}) \equiv C_R(t) + i C_I(t) \quad (14)$$

with $C_R(t)$ and $C_I(t)$ real functions of time. With this definition

$$C(t) = 2C_R(t) \cos(\omega_0 t) + 2C_I(t) \sin(\omega_0 t). \quad (15)$$

The emission profile is, then,

$$\begin{aligned} I(\omega) &= \frac{1}{\pi} \int_0^\infty dt \cos(\omega t) [2C_R(t) \cos(\omega_0 t) + 2C_I(t) \sin(\omega_0 t)] \\ &= \frac{1}{\pi} \int_0^\infty dt \{[\cos((\omega + \omega_0)t) + \cos((\omega - \omega_0)t)] C_R(t) \\ &\quad + [\sin((\omega + \omega_0)t) - \sin((\omega - \omega_0)t)] C_I(t)\}. \end{aligned} \quad (16)$$

Now we write $\omega = \omega_0 + \Delta\omega$, and calculate the spectrum around ω_0 . Moreover we ignore contributions to the spectrum of quantities such as $\int_0^\infty dt \cos((\omega + \omega_0)t) C_R(t)$ which would be significant if the functions $C_R(t)$ or $C_I(t)$ had temporal fluctuations in this frequency scale. Therefore we have

$$I(\Delta\omega) = \frac{1}{\pi} \int_0^\infty dt [\cos(\Delta\omega t) C_R(t) - \sin(\Delta\omega t) C_I(t)]. \quad (17)$$

3.1. Formalism in Liouville space

The cyclic invariance of the trace in (14) gives:

$$C_R(t) + i C_I(t) = \text{tr}(\mathbf{d}^+ \cdot U_u \mathbf{d} U_1^+). \quad (18)$$

We then define

$$\mathbf{d}(t) \equiv U_u(t) \mathbf{d} U_1^+(t), \quad (19)$$

and use Eqs. (11) to calculate the evolution of matrices $\mathbf{d}(t)$:

$$i\hbar \frac{d}{dt} \mathbf{d}(t) = H_u(t) \mathbf{d}(t) - \mathbf{d}(t) H_l(t). \quad (20)$$

Each element d_{ij} of matrix \mathbf{d} connects a state $|\psi_i^{(u)}\rangle$ belonging to the upper group with a state $|\psi_j^{(l)}\rangle$ of the lower group. It is useful to work in a Liouville space with states of the form $|i, j\rangle \equiv |\psi_i^{(u)}\rangle \otimes |\psi_j^{(l)}\rangle$. In this new space, matrices \mathbf{d} have vector structure and differential Eq. (20) can be written as:

$$i\hbar \frac{d}{dt} |\mathbf{d}(t)\rangle = \mathcal{L} |\mathbf{d}(t)\rangle, \quad (21)$$

where each matrix element $(\mathcal{L} \mathbf{d})_{ij}$ is calculated from

$$\begin{aligned} (\mathcal{L} \mathbf{d})_{ij} &= \sum_k H_{ik}^{(u)} d_{kj} - \sum_k d_{ik} H_{kj}^{(l)} \\ &= \sum_{n,m} (H_{in}^{(u)} \delta_{jm} - \delta_{in} H_{mj}^{(l)}) d_{nm}. \end{aligned} \quad (22)$$

More compactly, we can write

$$\mathcal{L} = H_u \otimes \mathbb{1} - \mathbb{1} \otimes H_l^t \quad (23)$$

where the upper index t denotes matrix transposition.

Within this formalism, the autocorrelation function can be written

$$C_R(t) + i C_I(t) = \langle \mathbf{d}(0) | \mathbf{d}(t) \rangle \quad (24)$$

with a ‘‘scalar product’’ of matrices A and B defined as

$$\langle A | B \rangle \equiv \text{tr}(A^+ B). \quad (25)$$

3.2. Characteristic time scales

There are three characteristic time scales in our problem. First, the typical correlation time of the perturbing field $\mathbf{E}(t)$, and in consequence, the characteristic time of $\mathcal{L}(t)$. This time scale is ruled by the kinetics of the charged particles in the plasma. Its order of magnitude is

$$\tau_c \simeq \frac{r_0}{v_0} \quad (26)$$

$$r_0 = \left(\frac{3}{4\pi N} \right)^{\frac{1}{3}} \quad (27)$$

$$v_0 = \sqrt{\frac{2kT}{m}} \quad (28)$$

where r_0 denotes the typical inter-particle distance, v_0 the mean quadratic velocity. N and T are the density and temperature of charged particles, and m their mass.

A second time scale is fixed by the correlations of the dipole-moment $\mathbf{d}(t)$. The spectral width is determined by this lifetime, τ_d , which is the true ‘‘unknown’’ of our problem.

Finally, a last time scale is fixed by characteristic values of \mathcal{L}/\hbar :

$$\frac{1}{\tau_H} \simeq \frac{1}{\hbar} \mathcal{L} \simeq \frac{1}{\hbar} q E_0 \frac{n^2 a_0}{Z}, \quad (29)$$

$$E_0 = \frac{q}{4\pi\epsilon_0 r_0^2}. \quad (30)$$

In the expressions above a_0 is the Bohr radius, n the principal quantum number of the upper group of states, Z the nuclear charge of the emitter and E_0 the typical local electric field. Evolution of the dipole moment $\mathbf{d}(t)$ would be fixed by this frequency scale if the perturbing electric field were static.

The relationship between τ_H and τ_c will determine the relevant physical phenomenon in spectral line broadening. When $\tau_c \gtrsim \tau_H$ we have ‘‘quasistatic’’ broadening, and both shape and width of the spectral line are fixed by the statistical distribution of the perturbing fields. Then we have $\tau_d \sim \tau_H$.

If $\tau_c \ll \tau_H$ the evolution of the dipole moment $\mathbf{d}(t)$ is much slower than the evolution of the perturbing fields and the correlation between $\mathcal{L}(t)$ and $\mathbf{d}(t)$ is quickly lost. In this case, perturbations are less efficient, since the emitter has enough time to ‘‘average’’ the electric field, and the time average of the field is considerably smaller than its statistical typical value. This regime is known as ‘‘impact broadening’’. In next section we will see that in this regime the width of the spectral line and therefore the lifetime $\tau_d \gg \tau_H \gg \tau_c$, is determined by the integral of the autocorrelation function of the perturbing field.

3.3. Impact approximation

The solution of Eq. (21) can be formally written as (the symbols $|\rangle$ are ignored for simplicity):

$$\mathbf{d}(t) = \mathbf{d}(t_0) + \frac{1}{i\hbar} \int_{t_0}^t dt' \mathcal{L}(t') \mathbf{d}(t') \quad (31)$$

$$\begin{aligned} &= \mathbf{d}(t_0) + \frac{1}{i\hbar} \int_{t_0}^t dt' \mathcal{L}(t') \mathbf{d}(t_0) \\ &\quad + \left(\frac{1}{i\hbar} \right)^2 \int_{t_0}^t dt' \mathcal{L}(t') \int_{t_0}^{t'} dt'' \mathcal{L}(t'') \mathbf{d}(t''). \end{aligned} \quad (32)$$

In order to calculate the spectral profile we need to average the correlation functions in Eq. (24):

$$\{C_R(t) + i C_I(t)\} = \text{tr}(\mathbf{d}^+(0) \cdot \{\mathbf{d}(t)\}), \quad (33)$$

so we have to calculate

$$\begin{aligned} \{\mathbf{d}(t)\} &= \{\mathbf{d}(t_0)\} + \frac{1}{i\hbar} \int_{t_0}^t \mathcal{L}(t') \mathbf{d}(t_0) \\ &+ \left(\frac{1}{i\hbar}\right)^2 \int_{t_0}^t dt' \int_{t_0}^{t'} \mathcal{L}(t') \mathcal{L}(t'') \mathbf{d}(t''). \end{aligned} \quad (34)$$

The average in the first integral satisfies $\{\mathcal{L}(t') \mathbf{d}(t_0)\} = \{\mathcal{L}(t')\} \{\mathbf{d}(t_0)\}$ since $\mathcal{L}(t')$ is posterior, and thus independent of $\mathbf{d}(t_0)$. Moreover, we are interested in the calculation of spectra for hydrogenlike emitters. For them, states in each group – upper and lower – are degenerate, and $H_{0u} = H_{0l} = 0$. Thus

$$\mathcal{L}(t') = q\mathbf{E}(t') \cdot (\mathbf{R}_u \otimes \mathbb{1} - \mathbb{1} \otimes \mathbf{R}_l^t) \quad (35)$$

and, consequently, $\{\mathcal{L}(t')\} = 0$.

We will use the arguments of the previous paragraph to calculate the double integral in (34). In the impact approximation ($\tau_c \ll \tau_d$) the dipole moment $\mathbf{d}(t)$ evolves much more slowly than $\mathcal{L}(t)$ and we can choose a time interval $\Delta t = t - t_0$ simultaneously satisfying the following two conditions:

$$\Delta t \ll \tau_d, \quad (36)$$

$$\Delta t \gg \tau_c. \quad (37)$$

In this case, the dipole moment $\mathbf{d}(t')$ is almost constant in the integration interval (t_0, t') of (34) and we have $\mathbf{d}(t') \approx \mathbf{d}(t_0)$. Thus, we can rewrite exp. (34) in the form:

$$\begin{aligned} \{\mathbf{d}(t_0 + \Delta t)\} - \{\mathbf{d}(t_0)\} \\ = \left(\frac{1}{i\hbar}\right)^2 \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \{\mathcal{L}(t') \mathcal{L}(t'')\} \{\mathbf{d}(t_0)\} \end{aligned} \quad (38)$$

since, as previously, t' and t'' are later than t_0 .

The perturbation $\mathcal{L}(t)$ felt by the emitter is a stationary random process, and thus the average $\{\mathcal{L}(t') \mathcal{L}(t'')\}$ only depends on the difference $\tau = t' - t''$. The last average is, precisely, the autocorrelation function of $\mathcal{L}(t)$ and can be written in terms of the autocorrelation function of the perturbing field (see exp. (35)):

$$\begin{aligned} \{\mathcal{L}(t') \mathcal{L}(t'')\} &= q^2 \sum_{i,j=x,y,z} \{E_i(t') E_j(t'')\} \\ &\times (R_i^{(u)} \otimes \mathbb{1} - \mathbb{1} \otimes R_i^{(l)}) (R_j^{(u)} \otimes \mathbb{1} - \mathbb{1} \otimes R_j^{(l)}) \\ &= \frac{q^2}{3} \{\mathbf{E}(t') \cdot \mathbf{E}(t'')\} (\mathbf{R}_u \otimes \mathbb{1} - \mathbb{1} \otimes \mathbf{R}_l^t)^2 \\ &\equiv \frac{q^2}{3} \Gamma(t' - t'') \mathcal{R}^2, \end{aligned} \quad (39)$$

where we have considered an isotropic system, so that $\{E_i(t') E_j(t'')\} = \frac{1}{3} \delta_{ij} \{\mathbf{E}(t') \cdot \mathbf{E}(t'')\}$.

Now we take the last result into (38). The integration range is shaded gray in Fig. 1. We change variables of integration to

$$\begin{aligned} \tau = t' - t''; \quad \zeta = t' + t''; \quad dt' dt'' = \frac{1}{2} d\tau d\zeta, \\ 0 \leq \tau \leq \Delta t, \quad \tau \leq \zeta \leq 2\Delta t - \tau. \end{aligned} \quad (40)$$

The region where the function $\Gamma(\tau)$ has significant values is shaded darker in Fig. 1. Integration over ζ gives rise to a factor Δt . Given that the range of the integration over τ includes all

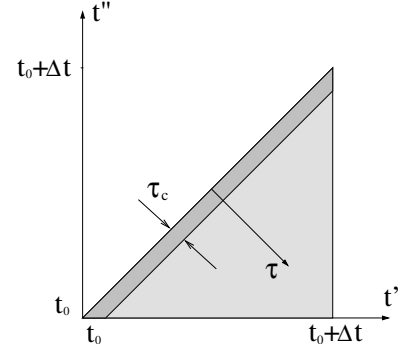


Fig. 1. Integration zone in exp. (38).

the relevant zone, we can change its upper limit to $+\infty$ (remember that $\Delta t = t - t_0 \gg \tau_c$). The double integral in (38), therefore, becomes

$$-\Delta t \Phi \cdot \{\mathbf{d}(t_0)\} \equiv \Delta t \left(\frac{q}{i\hbar}\right)^2 \int_0^\infty d\tau \Gamma(\tau) \frac{1}{3} \mathcal{R}^2 \{\mathbf{d}(t_0)\}, \quad (41)$$

where we have put

$$\Phi \equiv \left[\frac{1}{3} \left(\frac{q}{\hbar}\right)^2 \int_0^\infty d\tau \Gamma(\tau) \right] \mathcal{R}^2. \quad (42)$$

In this way, (38) may be written:

$$\frac{\{\mathbf{d}(t_0 + \Delta t)\} - \{\mathbf{d}(t_0)\}}{\Delta t} = -\Phi \cdot \{\mathbf{d}(t_0)\}. \quad (43)$$

Since $\Delta t \ll \tau_d$, this difference equation accounts for the rough sketch of the time derivative of $\mathbf{d}(t)$. Consequently, we can write:

$$\{\mathbf{d}(t)\} = \exp[-\Phi t] \mathbf{d}(0). \quad (44)$$

As Baranger wrote, (Baranger 1958b,c), “when the impact approximation is valid, it is allowable to replace the exact, fluctuating interaction between the atom and the perturbers by a constant effective interaction Hamiltonian” Φ .² The time behavior of the average of the dipole moment is determined both by the integral of the perturbing electric-field autocorrelation function and by the values of the \mathcal{R}^2 matrix elements. In general, for any transition it is convenient to diagonalize the matrix \mathcal{R}^2 and to write the vector operator $\mathbf{d}(0)$ in the eigenbase of \mathcal{R}^2 . When this is done, the autocorrelation function becomes

$$\{C_R(t) + i C_I(t)\} = \sum_k e^{-\phi_k t} \langle k | \mathbf{d}^+(0) \cdot \mathbf{d}(0) | k \rangle, \quad (45)$$

where the summation covers all eigenstates $|k\rangle$ of \mathcal{R}^2 with eigenvalues ϕ_k . The corresponding matrix element accounts for the intensity of that spectral component. But in the case of the hydrogen atom everything is easier. For hydrogen, the vector $\mathbf{d}(0)$ is an eigenvector of \mathcal{R}^2 , that is (in atomic units)

$$\mathcal{R}^2 \mathbf{d} = \phi_{m'l'} \mathbf{d}, \quad (46)$$

being

$$\phi_{m'l'} = \frac{9}{4} \left[(n^2 - n'^2)^2 - (n^2 + n'^2) \right]. \quad (47)$$

² The characteristic values of Φ give the width of the spectral line, which has a Lorentzian shape. The value of the integral of function $\Gamma(\tau)$ is proportional to $E_0^2 \tau_c$, E_0 being the typical perturber fields and τ_c the time of correlation loss for that field. These magnitudes are $E_0 \sim N^{2/3}$ and $\tau_c \sim N^{-1/3} T^{-1/2}$ so that the line width is proportional to $N T^{-1/2}$.

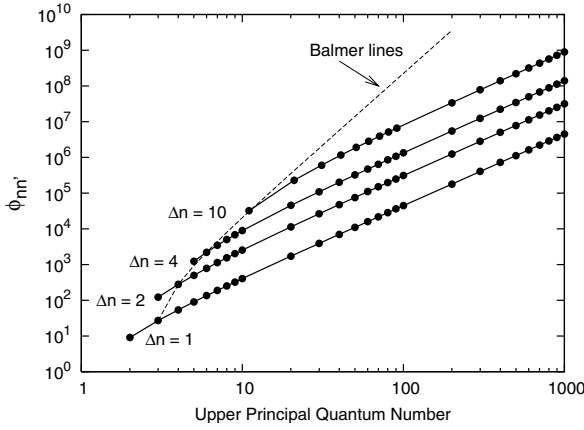


Fig. 2. Stark width parameter depending on the upper group principal quantum number. The values $\phi_{nn'}$ have been represented as a function of n with $n' = n - \Delta n$ given as pointed out in the figure. The broken line gives the values corresponding to the Balmer series, in which $n' = 2$. The trend for fixed Δn is $\sim n^2$, while for fixed n' , $\phi_{nn'}$ changes as n^4 .

(n and n' are the principal quantum numbers for the upper and the lower group respectively). The proof of this result is shown in Appendix A.

Under these conditions, expression (45) becomes simpler:

$$\{C_R(t)\} = e^{-\phi t}, \quad \{C_I(t)\} = 0, \quad (48)$$

$$\phi = \phi_{nn'} \frac{1}{3} \left(\frac{qa_0}{Z\hbar} \right)^2 \int_0^\infty d\tau \Gamma(\tau), \quad (49)$$

with a_0 the Bohr radius and Z the emitter nuclear charge. (We have normalized the function so that $C_R(0) = \text{tr}[\mathbf{d}^+ \cdot \mathbf{d}] = 1$).

The particular case with $n = n' + 1$ is interesting. Then

$$\phi_{n+1 n} = \frac{9}{2} n(n+1). \quad (50)$$

Compare this expression with the approximate expression labelled as (27) in Griem (1967), and see Fig. 2, where the dependency of impact width upon the principal quantum number of the upper level is shown for different values of Δn .

4. Results and discussion

The former section gave the analysis of the Stark broadening term in the frame of the impact approximation completely excluding slow varying fields. It is necessary, then, to consider that collisions are weak and rapid. When a strong collision takes place – when a perturber passes very close to the emitter atom – the resulting phase change in the evolution operator cannot be considered very small. As a consequence, the approximation allowing us to pass from expression (34) to (38) cannot be considered valid and these collisions must be excluded from our calculation. Of course there are few such events, but the so-called “*strong collisions*” must be taken into account. These collisions represented by the first term in (1) have not been considered here. The most adequate treatment for these cases is the original one from Lorentz that correctly considers that those collisions are all independent, never overlap in time and that, when they happen the effect of the rest of the perturbers is completely negligible. These collisions may be considered to be independent of the weak collisions that give rise to the broadening that is being studied here. Therefore, their effect may be taken into account simply adding a term to the result of the broadening due

to weak collisions, as is done in expression (1). The analysis of those cases is beyond the scope of this work.

Slowly varying fields due to ionic perturbers have not been considered in our study. Such fields with correlation time $\tau_c \gtrsim \tau_H$, or $\tau_c \gtrsim \tau_d$ induce “quasistatic broadening”. For this to happen the ionic density must be high enough. However, when the ion density is very low, ions can no longer be considered static during the typical dipole relaxation time. As already pointed out by Griem (2005) – see also Griem (1967, 1974) –, this happens, for example, for the conditions of radio recombination lines studied in Bell et al. (2000), where ions effect can be considered as “impact”. In those cases, then, the dominant broadening is ionic impact, as their fields, as they are slower, have a correlation integral (42) larger than that of electrons³.

The treatment considered in the previous section differs from the standard treatment by a fundamental point. Usually, in the traditional treatment, the statistics over the perturbation undergone by the emitter is calculated considering the quantities that characterize the collisions, i.e. the velocity and impact parameter of the individual perturbers. The treatment followed here does not consider how to perform such averages as this is unnecessary here. Statistics over particle perturbation conditions is considered in an average over the perturber field correlation function. Whatever the value of such a function, the proportions between the broadenings of the different hydrogen spectral lines are not affected provided the impact approximation is valid.

However, there is a problem in the calculation of the perturber electric field correlation function $\Gamma(t)$ appearing first in exp. (39). When one studies a plasma formed by independent particles – as is usually done in the analytical simplifications of Stark broadening – that function diverges when $t \rightarrow 0^+$. This divergence is a consequence of the field divergence for very short distances. In this way, the average appearing in expression (39) must be determined carefully, excluding the configurations in which a perturber is very close to the emitter. Therefore the limitation mentioned in the first paragraph is common to both the present and the traditional formalism. In this last one, the problem is solved considering a lower integration limit for the impact parameters of the perturbers and including the effect of very

³ In this work we have been calling “impact approximation” the process allowing us to derive (38) from Eq. (34). That is, it is necessary that the average that appears in the second integral in (34) can be written as it appears in (38). This requires that the dipole correlation time is much higher than the perturber field typical fluctuation time. However, the term “impact approximation” is usually employed with the meaning used in the first works on Stark broadening that also include the elastic collision approximation. These two approximations are of course independent. The Liouville operator in Eq. (34) may, or may not, include inelastic collisions, which does not affect the argument used in the derivation of Eq. (38) from Eq. (34). In order to follow the usual denomination we could substitute along all our work the term “impact approximation” by perturber field “fast fluctuation approximation”. In particular, when it is said that impact broadening is the dominant effect for radio recombination lines we possibly need to include inelastic collisions but we can keep the frame of fast fluctuations. Of course, the expression obtained here for the width operator is valid only when one only considers elastic collisions.

⁴ If one considers free and independent particles with straight line trajectories as in standard theory, $\Gamma(t) \sim 1/t$. This gives rise to a logarithmic divergence at both limits. For very long times this divergence is trivially corrected using a cutting radius (such as the Debye radius) given that real trajectories are affected by the other perturbers. For short distances the correction of the minimum radius must be introduced to separate the effects of strong and weak collisions, as already mentioned before.

close collisions through a strong collision term in the frame of the Lorentz model.

In anycase, we insist, the limitations of impact broadening may affect the magnitude accuracy of the integral appearing in (49), but would not affect expression (47), i.e. the relationship between the Stark width and the principal quantum numbers of the levels involved in the transition under study that is what we are considering in this work.

Expressions (46) and (47) show that Stark widths of the individual components of the transition are independent of the angular momentum quantum numbers involved. All the components are equally broadened. This is a consequence of the symmetry properties of the hydrogen atom, and, in general, one cannot expect it to happen for other elements. Appendix A shows that this homogeneity is due to the so-called “*interference term*” in the width operator. If this term did not appear, the width of each component arising from a state (n, l, m) to another state (n', l', m') would have a width proportional to

$$\frac{9}{4} \left[(n^4 - n'^2) + (n'^4 - n^2) - n^2 l(l+1) - n'^2 l'(l'+1) \right]. \quad (51)$$

There is still one thing remaining. More than thirty years ago a discussion took place about the physical meaning of the so-called “*interference term*” (Hey & Griem 1975; Voslamber 1976; Griem & Hey 1976). However, that term does not account for any physical requirements: it results from a mathematical development in series of powers. It is a consequence of the fact that the states of the upper group and those of the lower group “feel” the same perturbing field. This keeps coherence in the evolution of those states and reduces the broadening effect due to the collisions. In our study, transitions due to the collisions between the states of the upper group and the states of the lower group have not been taken into account. This is the main limitation of this treatment, which ignores inelastic collisions that, for many conditions, are dominant (Griem 2005). Accounting for such collisions is beyond our objective in this work, which is to give a simple and compact expression for the Stark width of hydrogen lines when both conditions are fulfilled: impact broadening for weak collisions in the frame of the “no-quenching” approximation.

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Appendix A: Calculation of the width operator

Here we prove expression (46), reproduced below

$$\mathcal{R}^2 |d\rangle = \phi_{nn'} |d\rangle \quad (A.1)$$

or, equivalently,

$$R_n^2 d + d R_{n'}^2 - 2 R_n d R_{n'} = \phi_{nn'} d \quad (A.2)$$

(in this expression we have written

$$R_n d R_{n'} \equiv \sum_{i=x,y,z} R_i^{(n)} d R_i^{(n')} \quad (A.3)$$

that corresponds to the so-called “*interference term*”).

The proof of exp. (A.2) involves two parts. First, the two first addends that depend on the squares of \mathbf{R} operator in each of the groups of states with principal quantum number n are evaluated. Second, the interference term is obtained.

A.1. First part

The hydrogen atom has a special symmetry that gives rise to accidental degeneracy. Such symmetry leads to commutation between the so-called Runge-Lenz vector, \mathbf{A} , and the atom Hamiltonian. That vectorial operator – properly normalized – forms, together with the angular momentum operator, \mathbf{L} , a set of generators for the $SO(4)$ group that may be separated in the tensor product of two $SO(3)$ groups. The generators of each of these groups are vector operators defined using the expressions :

$$\mathbf{J}(1) = \frac{1}{2} (\mathbf{L} + \mathbf{A}), \quad \mathbf{J}(2) = \frac{1}{2} (\mathbf{L} - \mathbf{A}). \quad (A.4)$$

These operators commute between them and their commutation rules correspond to those of an angular momentum (Biedenharn & Louck 1981):

$$[J_i(a), J_j(a)] = i \varepsilon_{ijk} J_k(a), \quad i, j, k = x, y, z; \quad a = 1, 2. \quad (A.5)$$

All these matrixes commute with the hydrogen atom Hamiltonian. In addition, in each group of states with principal quantum number n , operators $J^2(1)$ and $J^2(2)$ are diagonal matrixes that are proportional to unity with eigenvalues (Biedenharn & Louck 1981):

$$J_n^2(1) = J_n^2(2) = \frac{n^2 - 1}{4}. \quad (A.6)$$

In Quantum Mechanics, as in Classical Mechanics, the Runge-Lenz vector is perpendicular to angular momentum, i.e. $\mathbf{L} \cdot \mathbf{A} = 0$ (Biedenharn & Louck 1981).

From these properties, we can write

$$\begin{aligned} A^2 &= J^2(1) + J^2(2) - 2\mathbf{J}(1) \cdot \mathbf{J}(2), \\ L^2 &= J^2(1) + J^2(2) + 2\mathbf{J}(1) \cdot \mathbf{J}(2). \end{aligned} \quad (A.7)$$

Therefore, for each subspace of states with principal quantum number n it can be written

$$A_n^2 = n^2 - 1 - L_n^2. \quad (A.8)$$

The Runge-Lenz vector is interesting as it is related to the \mathbf{R} operator that appears in expression (A.2). To be exact, in the subspace of states with principal quantum number n one has (Demkov et al. 1969)

$$\mathbf{R}_n = \frac{3}{2} n \mathbf{A}_n. \quad (A.9)$$

Then,

$$R_n^2 = \frac{9}{4} n^2 (n^2 - 1 - L_n^2) \quad (A.10)$$

in the spherical basis $|n, l, m\rangle$, each matrix element of each of the two first operators in (A.2) are written simply:

$$\begin{aligned} \langle n, l, m | R_n^2 | n', l', m' \rangle &= \frac{9}{4} n^2 (n^2 - 1 - l(l+1)) \\ &\quad \times \langle n, l, m | d | n', l', m' \rangle \end{aligned} \quad (A.11)$$

$$\begin{aligned} \langle n, l, m | d R_{n'}^2 | n', l', m' \rangle &= \frac{9}{4} n'^2 (n'^2 - 1 - l'(l'+1)) \\ &\quad \times \langle n, l, m | d | n', l', m' \rangle. \end{aligned} \quad (A.12)$$

A.2. Second part

For the interference term there is more algebra. Work is done in the basis $|n, l, m\rangle$ of eigenstates of H_0 , L^2 and L_z . It is convenient to work with vector operators using their spherical components $-1, 0, +1$ instead of using the Cartesian basis x, y, z . We calculate, then, each matrix element of the \bar{M} -th component of the vector operator appearing in (A.3) using the expression

$$\begin{aligned} \langle n, l, m | V_{\bar{M}} | n', l', m' \rangle &\equiv \langle n, l, m | \mathbf{R}_n D_{\bar{M}} \mathbf{R}_{n'} | n', l', m' \rangle \\ &= \sum_M \sum_{l_1, m_1} \sum_{l_2, m_2} (-1)^M \langle n, l, m | \mathbf{R}_M | n, l_1, m_1 \rangle \\ &\quad \times \langle n, l_1, m_1 | D_{\bar{M}} | n', l_2, m_2 \rangle \\ &\quad \times \langle n', l_2, m_2 | \mathbf{R}_{-M} | n', l', m' \rangle. \end{aligned} \quad (\text{A.13})$$

We use the Wigner-Eckart theorem to write, for each of the three vector operators appearing in (A.13) (Biedenharn & Louck 1981),

$$\langle n, l, m | \mathbf{R}_M | n, l_1, m_1 \rangle = C_{m_1 M m}^{l_1 1 l} \langle n, l | \mathbf{R} | n, l_1 \rangle \quad (\text{A.14})$$

where $C_{\alpha\beta\gamma}^{abc}$ is a Clebsch-Gordan coefficient and $\langle n, l | \mathbf{R} | n, l_1 \rangle$ is the corresponding reduced matrix element. Expression (A.13) can be rewritten:

$$\begin{aligned} \langle n, l, m | V_{\bar{M}} | n', l', m' \rangle &= \sum_{l_1, l_2} \langle n, l | \mathbf{R} | n, l_1 \rangle \langle n, l_1 | \mathbf{d} | n', l_2 \rangle \\ &\quad \times \langle n', l_2 | \mathbf{R} | n', l' \rangle \\ &\quad \times \sum_{M, m_1, m_2} (-1)^M C_{m_1 M m}^{l_1 1 l} C_{m_2 M m_1}^{l_2 1 l_1} C_{m' - M m_2}^{l' 1 l_2}. \end{aligned} \quad (\text{A.15})$$

The result of this last summation of products of three Clebsch-Gordan coefficients is known. For example, in expression labeled as (18) on page 260 of Varshalovich et al. (1988) it is written:

$$\begin{aligned} \sum_{\alpha\beta\gamma} (-1)^{b+\beta} C_{-\beta\gamma\alpha}^{b \ c \ a} C_{\delta\beta\epsilon}^{d \ b \ e} C_{\alpha\phi\delta}^{c \ a \ f} &= (-1)^{b+c+d+f} \\ &\quad \times \sqrt{(2a+1)(2d+1)} C_{\gamma\phi\epsilon}^{c \ f \ e} \begin{Bmatrix} abc \\ efd \end{Bmatrix}. \end{aligned} \quad (\text{A.16})$$

Using the symmetry relation

$$C_{\alpha\beta\gamma}^{abc} = (-1)^{a+b-c} C_{\beta\alpha\gamma}^{bac} \quad (\text{A.17})$$

applied to the last Clebsch-Gordan member of the summation, one finds that exp. (A.15) may be written as:

$$\begin{aligned} \langle n, l, m | V_{\bar{M}} | n', l', m' \rangle &= \sum_{l_1, l_2} \langle n, l | \mathbf{R} | n, l_1 \rangle \langle n, l_1 | \mathbf{d} | n', l_2 \rangle \\ &\quad \times \langle n', l_2 | \mathbf{R} | n', l' \rangle (-1)^{l_1+l_2} \sqrt{2l_1+1} \sqrt{2l_2+1} C_{m' \bar{M} m}^{l' 1 l} \begin{Bmatrix} l_2 1 l' \\ l 1 l_1 \end{Bmatrix} \\ &\equiv C_{m' \bar{M} m}^{l' 1 l} \langle n, l | \mathbf{V} | n', l' \rangle. \end{aligned} \quad (\text{A.18})$$

The triangular rules of the $6j$ element that appears in this expression force $|l - l'| \leq 1$ (which cannot be different, because operator \mathbf{V} is a vector). This condition, together with the selection rules for operator \mathbf{R} , limit the possible cases of values l_1 and l_2 to those given in Table (A.1). In that same table the values of the corresponding $6j$ element are shown. These were obtained from the particular expressions (Brink & Satchler 1993)

$$\begin{Bmatrix} a 1 a + 1 \\ a 1 a + 1 \end{Bmatrix} = \begin{Bmatrix} a + 11 a \\ a + 11 a \end{Bmatrix} = \frac{1}{(2a+1)(a+1)(2a+3)} \quad (\text{A.19})$$

$$\begin{Bmatrix} a \ 1 \ a + 1 \\ a + 21 a + 1 \end{Bmatrix} = \begin{Bmatrix} a + 21 a + 1 \\ a \ 1 \ a + 1 \end{Bmatrix} = \frac{1}{(2a+3)}. \quad (\text{A.20})$$

Table A.1. Possible cases of values for l_1 and l_2 in exp. (A.18) as a function of the extreme values l and l' . The last column gives the value of the corresponding $6j$ element according to last expressions (A.19) and (A.20).

l	l_1	l_2	l'	$\begin{Bmatrix} l_2 1 l' \\ l 1 l_1 \end{Bmatrix}$
l	$l+1$	$l+2$	$l+1$	$1/(2l+3)$
l	$l+1$	l	$l+1$	$1/[(2l+1)(l+1)(2l+3)]$
l	$l+1$	l	$l-1$	$1/(2l+1)$
l	$l-1$	l	$l+1$	$1/(2l+1)$
l	$l-1$	l	$l-1$	$1/[(2l-1)l(2l+1)]$
l	$l-1$	$l-2$	$l-1$	$1/(2l-1)$

In order to develop expression (A.18) it is convenient to write the reduced matrix elements appearing in it according to the radial integrals of the hydrogen atom. Then one has (Bethe & Salpeter 1957)

$$\langle n_1, l | \mathbf{R} | n_2, l+1 \rangle = -\sqrt{\frac{l+1}{2l+1}} R_{n_2 l+1}^{n_1 l} \quad (\text{A.21})$$

$$\langle n_1, l | \mathbf{R} | n_2, l-1 \rangle = \sqrt{\frac{l}{2l+1}} R_{n_2 l-1}^{n_1 l} \quad (\text{A.22})$$

and using

$$R_{n l-1}^{n l} = R_{n l}^{n l-1} = \frac{3}{2} n^2 l A_{n l} \quad (\text{A.23})$$

being

$$A_{n l} = \frac{1}{n l} \sqrt{n^2 - l^2}. \quad (\text{A.24})$$

We may develop expression (A.18) for the case $l' = l+1$. The case $l' = l-1$ may be obtained from our result applying the formal change $n \leftrightarrow n'$ and $l \leftrightarrow (l-1)$. One obtains, then,

$$\begin{aligned} \langle n, l | \mathbf{V} | n', l+1 \rangle &= \sum_{l_1, l_2} \langle n, l | \mathbf{R} | n, l_1 \rangle \langle n, l_1 | \mathbf{d} | n', l_2 \rangle \\ &\quad \times \langle n', l_2 | \mathbf{R} | n', l+1 \rangle (-1)^{l_1+l_2} \sqrt{2l_1+1} \sqrt{2l_2+1} \begin{Bmatrix} l_2 1 l+1 \\ l 1 l_1 \end{Bmatrix} \\ &= -\sqrt{\frac{l+1}{2l+1}} \left[\frac{l+2}{2l+3} R_{n l+1}^{n l} R_{n' l+2}^{n' l+1} R_{n' l+1}^{n' l+2} \right. \\ &\quad \left. + \frac{1}{(2l+1)(2l+3)} R_{n l+1}^{n l} R_{n' l}^{n' l+1} R_{n' l+1}^{n' l} + \frac{l}{2l+1} R_{n l-1}^{n l} R_{n' l}^{n' l-1} R_{n' l+1}^{n' l} \right]. \end{aligned} \quad (\text{A.25})$$

We use, now, (A.23) and (A.24):

$$\begin{aligned} \langle n, l | \mathbf{V} | n', l+1 \rangle &= -\frac{9}{4} n^2 n'^2 \sqrt{\frac{l+1}{2l+1}} \frac{l+1}{(2l+1)(2l+3)} \\ &\quad \times \left[(l+2)^2 (2l+1) A_{n l+1} A_{n' l+2} R_{n' l+2}^{n' l+1} \right. \\ &\quad \left. + A_{n l+1} A_{n' l+1} R_{n' l}^{n' l+1} + l^2 (2l+3) A_{n l} A_{n' l+1} R_{n' l}^{n' l+1} \right]. \end{aligned} \quad (\text{A.26})$$

Now we use the recurrence relations for the radial integrals obtained by Infeld & Hull (1951) (see also Hey (2006)), that are shown here:

$$2\ell A_{n' \ell} R_{n' \ell-1}^{n' \ell} = (2\ell+1) A_{n' \ell+1} R_{n' \ell}^{n' \ell+1} + A_{n' \ell+1} R_{n' \ell+1}^{n' \ell}, \quad (\text{A.27})$$

$$2\ell A_{n' \ell} R_{n' \ell-1}^{n' \ell-1} = (2\ell+1) A_{n' \ell+1} R_{n' \ell}^{n' \ell+1} + A_{n' \ell+1} R_{n' \ell+1}^{n' \ell+1}. \quad (\text{A.28})$$

(expression (A.28) may be obtained from (A.27) replacing n by n' and taking into account the symmetry of radial integrals

with regard to an exchange of upper and lower indexes). In order to obtain a new useful relation between these functions, one may multiply equality (A.28) by $(2\ell + 1)$, subtract it from (A.27) and substitute the value ℓ by $(\ell - 1)$ in the result. One obtains:

$$2\ell A_{n'\ell} R_{n'\ell}^{n\ell-1} = (2\ell - 1)A_{n'\ell-1} R_{n'\ell-1}^{n\ell-2} - A_{n'\ell-1} R_{n'\ell-2}^{n\ell-1}. \quad (\text{A.29})$$

With these relations we transform the terms inside $\left[\right]$ in expression (A.26). We write

$$\begin{aligned} \left[\right] &= (l+2)(2l+1)A_{n'l+1} \frac{1}{2} \left[(2l+3)A_{n'l+1} R_{n'l+1}^{nl} - A_{n'l+1} R_{n'l}^{n'l+1} \right] \\ &+ (l+1)A_{n'l+1} A_{n'l+1} R_{n'l}^{n'l+1} \\ &+ l(2l+3)A_{n'l+1} \frac{1}{2} \left[(2l+1)A_{n'l+1} R_{n'l+1}^{nl} + A_{n'l+1} R_{n'l}^{n'l+1} \right]. \quad (\text{A.30}) \end{aligned}$$

The first line in this expression has been obtained substituting the product $(l+2)A_{n'l+2} R_{n'l+2}^{n'l+1}$ using (A.29) with $\ell = l+2$. The third line in (A.30) is obtained substituting the product $lA_{n'l+1} R_{n'l}^{n'l+1}$ using (A.28) with $\ell = l$.

Grouping together terms in (A.30):

$$\begin{aligned} \left[\right] &= \left[\frac{1}{2}(l+2)(2l+1)(2l+3) \right] (A_{n'l+1})^2 R_{n'l+1}^{nl} \\ &+ \left[\frac{1}{2}l(2l+1)(2l+3) \right] (A_{n'l+1})^2 R_{n'l}^{n'l+1} \\ &+ \left[\frac{1}{2}l(2l+3) + (l+1) + \frac{1}{2}(l+2)(2l+1) \right] \\ &\times A_{n'l+1} A_{n'l+1} R_{n'l}^{n'l+1} \\ &= \frac{1}{2}(2l+1)(2l+3) R_{n'l+1}^{nl} \left\{ (l+1) \left[(A_{n'l+1})^2 + (A_{n'l+1})^2 \right] \right. \\ &\left. + \left[(A_{n'l+1})^2 - (A_{n'l+1})^2 \right] \right\}. \quad (\text{A.31}) \end{aligned}$$

Now one can use (A.24) to write

$$\begin{aligned} \left[\right] &= \frac{(2l+1)(2l+3)}{2(l+1)} R_{n'l+1}^{nl} \left[\frac{n^2 - (l+1)^2}{n^2} + \frac{n'^2 - (l+1)^2}{n'^2} \right. \\ &\left. + \frac{n^2 - (l+1)^2}{n^2(l+1)} - \frac{n'^2 - (l+1)^2}{n'^2(l+1)} \right] \\ &= \frac{1}{2n^2 n'^2} \frac{(2l+1)(2l+3)}{l+1} R_{n'l+1}^{nl} \\ &\times \left[2n^2 n'^2 - (l+1)^2(n^2 + n'^2) + (n^2 - n'^2)(l+1) \right] \\ &= \frac{1}{2n^2 n'^2} \frac{(2l+1)(2l+3)}{l+1} R_{n'l+1}^{nl} \\ &\times \left[2n^2 n'^2 - n^2 l(l+1) - n'^2 l(l+1)(l+2) \right]. \quad (\text{A.32}) \end{aligned}$$

If we now take this result to (A.26):

$$\begin{aligned} \langle n, l \| \mathbf{V} \| n', l+1 \rangle &= -\frac{9}{8} \sqrt{\frac{l+1}{2l+1}} R_{n'l+1}^{nl} \\ &\times \left[2n^2 n'^2 - n^2 l(l+1) - n'^2 l(l+1)(l+2) \right]. \quad (\text{A.33}) \end{aligned}$$

But, according to exp. (A.21),

$$\begin{aligned} \langle n, l \| \mathbf{V} \| n', l+1 \rangle &= \frac{9}{8} \left[2n^2 n'^2 - n^2 l(l+1) - n'^2 l(l+1)(l+2) \right] \\ &\times \langle n, l \| \mathbf{R} \| n', l+1 \rangle. \quad (\text{A.34}) \end{aligned}$$

As has been mentioned, this relation equally may be found if one studies the change from l to $l' = l - 1$. Finally, taking into account (A.13), (A.18) and (A.34), one can write:

$$\begin{aligned} \langle n, l, m \| \mathbf{R}_n \mathbf{d} \mathbf{R}_{n'} \| n', l', m' \rangle &= \frac{9}{8} \left[2n^2 n'^2 - n^2 l(l+1) - n'^2 l'(l'+1) \right] \\ &\times \langle n, l, m \| \mathbf{d} \| n', l', m' \rangle. \quad (\text{A.35}) \end{aligned}$$

A.3. Third part

We must only group together terms. Joining expressions (A.11), (A.12) and (A.35) one obtains:

$$\begin{aligned} \langle n, l, m \| \mathbf{R}_n^2 \mathbf{d} + \mathbf{d} \mathbf{R}_{n'}^2 - 2\mathbf{R}_n \mathbf{d} \mathbf{R}_{n'} \| n', l', m' \rangle &= \frac{9}{4} \left[(n^2 - n'^2)^2 \right. \\ &\left. - (n^2 + n'^2) \right] \langle n, l, m \| \mathbf{d} \| n', l', m' \rangle \quad (\text{A.36}) \end{aligned}$$

leading to expressions (A.1) and (47).

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