

Classical-Path Calculation of the S Matrices for Stark Broadening of the Lyman- α Line

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The S matrices for Stark broadening of the $n=1$ and $n=2$ levels of hydrogen based on the straight-line classical-path model are calculated numerically. The interaction terms (excluding exchange) are included to all orders, and time ordering of the operators in the S matrix is retained. The resultant values for the impact-broadening operator Φ are presented. These results are compared with previous approximations based on the same model. The effect of the present calculations on the center and near wings of the Lyman- α line profile is considered.

I. INTRODUCTION

In the calculations of the Stark broadened line, profiles for the hydrogen lines performed by Griem *et al.*¹ (hereafter referred to as GKSI) an approximate solution of the equation for the time-development operator in the interaction representation was obtained by keeping the first nonzero (second-order) term in the iterative solution (Dyson expansion). Only the dipole term in the free-electron atom interaction energy was used. This dipole term was calculated using the straight-line classical-path model. In order to determine the electron impact-broadening operator [Eq. (1)] integration over the electron impact parameter ρ was required. In performing this integration, divergences were encountered at both zero and infinite impact parameters. The divergence at large impact parameters can be removed by cutting off the integral at a distance of the order of the Debye length. In GKSI, the divergence as ρ approached zero was removed by cutting off the integral at a minimum impact parameter ρ_{\min} . This ρ_{\min} was found by setting the first nonzero term of the S matrix equal to unity, corresponding to the breakdown of the perturbation expansion. In general, ρ_{\min} varies from term to term of the S matrix. For convenience a "mean" value for ρ_{\min} was chosen. An estimate was made for the strong collision contribution in the calculations of the H_β profile by Griem, Kolb, and Shen² (GKSII), and the calculations of the H_α and H_γ lines by Bacon and Edwards.^{3,4} Griem⁵ summed the first ten terms of the Dyson series for the Lyman- α line in an approximation in which the interaction potential commutes with itself at different times. The latter author also estimated

the contribution to Φ from the quadrupole term in the interaction energy. Recently, Shen and Cooper⁶ (SC) were able to make an approximation (similar to that of Griem⁵) by which the S matrix could be summed to all orders, yielding a classical-path S matrix which no longer diverged in the limit of zero impact parameter. The technique was illustrated by calculating the Φ matrix elements for Lyman α .

The purpose of this paper is not to present the best possible Lyman- α profiles but rather to compare, in what we believe to be the most concise way, the Φ operator calculated by including all terms in the multipole interaction and retaining the correct time ordering with the result of the previous calculations outlined above. With this in mind, we fix the upper cutoff of the impact parameter at the Debye length. The inclusion of the approximate effects of level splittings, caused by the ion fields, and the finite duration of the collisions, as was done in the calculations of Kepple and Griem,⁷ are neglected. Modifications suggested by Chappell, Cooper, and Smith⁸ are also neglected. These modifications which affect the upper cutoff will not affect the results reported here insofar as the constants K_1 and K_2 [Eqs. (9) and Table I] are concerned.

II. THEORY AND NOTATION

In the Stark broadening theories of Kolb and Griem⁹ and Baranger¹⁰ the impact broadening operator Φ , which contains the effects of the free electrons on the Stark broadened profiles of spectral lines, is given by

$$\Phi = 2\pi N \int v f(v) dv \int \rho d\rho \{S_{n n'}^* S_{n' n} - 1\}_{av}. \quad (1)$$

TABLE I. Values of K_1 and K_2 determined by this calculation compared to the values of Griem (Ref. 5). The values of K_1 and K_2 determined by other approximations are independent of temperature and are given below. GKS I: $K_1=K_2=0.0$. GKS II: $K_1=K_2=1.0$. SC: $K_1=K_2=0.579$. Dipole-only: $K_1=-0.390$, $K_2=1.62$.

Temperature ($^{\circ}$ K)	K_1	K_2	av K $\frac{1}{4}(3K_2+K_1)$	Griem (1965) $K_1=K_2$ $(0.582+3kT/3E_H)$
5×10^3	-0.390	1.62	1.12	0.666
1×10^4	-0.405	1.61	1.11	0.751
2×10^4	-0.413	1.58	1.08	0.920
3×10^4	-0.421	1.48	1.01	1.089
4×10^4	-0.430	1.40	0.94	1.258
5×10^4	-0.445	1.33	0.89	1.427
6×10^4	-0.455	1.25	0.83	1.596

N is the electron density, v is the electron velocity, $f(v)$ is the normalized Maxwellian velocity-distribution function, and ρ is the impact parameter. In the classical-path approximation $\{ \}_{\text{av}}$ signifies an average over the directions of the impact parameters and velocities. S_n and $S_{n'}$ are the S matrices for a single encounter, with closest approach occurring at $t=0$ for the upper levels (principal quantum number n), and lower levels (principal quantum number n'), respectively. The S matrix is given by

$$S = U(\infty, -\infty), \quad (2)$$

where U is the time-development operator in the interaction representation. S is found for the upper and lower levels by integrating

$$i\hbar \frac{\partial U}{\partial t} = \tilde{V}(t)U \quad (3)$$

from $-\infty$ to $+\infty$. As far as line radiation for transitions between states with small principal quantum numbers is concerned; we are only interested in the matrix elements of U between states of the same principal quantum number (no quenching assumption).

In the straight-line classical-path model,

$$\tilde{V}(t) = \exp[iH_0 t/\hbar] V(t) \exp[-iH_0 t/\hbar], \quad (4)$$

where $V(t)$ is the classical interaction energy between a free electron and a radiating atom. H_0 is the Hamiltonian of the atomic system. We make the usual assumption (see GKS I¹ and SC,⁶ for example) that for the degenerate hydrogen system the effect of level splittings caused by the ion fields may be neglected, and we may set the exponentials in Eq. (4) equal to unity. In spite of setting the exponential in Eq. (4) equal to unity so that $\tilde{V}(t) = V(t)$, $V(t)$ does not commute with itself at different times, since implicitly we are only taking matrix

elements within a given principal quantum number (see Smith, Vidal, and Cooper¹¹). SC and Griem⁵ assumed that they did commute. It should be noted that the effects of level splitting caused by the ion fields, together with the other effect mentioned in Sec. I may be treated in an approximate manner, by first assuming that the exponentials in Eq. (4) may be replaced by unity and then applying the cutoff procedures given by Kepple and Griem.⁷ Such considerations are beyond the scope of this paper and will not be considered further.

Substituting $V(t)$ for $\tilde{V}(t)$, and using the spherical wave functions $|nlm\rangle$ as bases, we may write Eq. (3), in a. u., as

$$\frac{i\partial}{\partial t} \langle nlm | U | n'l'm' \rangle = \sum_{l''m''i} \langle nlm | V^i | n'l''m'' \rangle \times \langle n'l''m'' | U | n'l'm' \rangle, \quad (5)$$

Note that when the differential equations of Eq. (5) are solved, the full time ordering of the iterative (Dyson series) solution is retained. The sum over the index i on the right-hand side of Eq. (5) indicates a sum over i -pole terms in the multipole expansion. The general expression for the V 's is given in the Appendix.

Evaluation of the impact-broadening operator Φ for a particular hydrogen line caused by transitions from states with principal quantum number n to states with principal quantum number n' involves two sets of coupled equations (5); one set for n and the other for n' . In Sec. III, we consider the Lyman- α line.

III. APPLICATION TO LYMAN- α LINE

The Lyman- α line involves the transition from states with principal quantum number $n=2$ to states with principal quantum number $n'=1$. The

relevant nonzero V^i matrix elements are given in the Appendix. The only nonzero term in the case of $n'=1$ level is the monopole term $\langle 100 | V^0 | 100 \rangle = \exp(-2R)(1+1/R)$, where $R = |\vec{\rho} + \vec{v}t|$. (a.u. are used throughout unless otherwise stated.) Since the $n'=1$ state consists of only one level ($|100\rangle$), the evaluation of the S matrix involves a set of two coupled equations. (U has a real and imaginary part.) The integration of Eq. (5) for $n'=1$ was carried out numerically. The real part remained essentially one and the imaginary part zero, thus confirming that the usual assumption of an unperturbed $n'=1$ state is good.

For Lyman- α , Eq. (1) may therefore be written

$$\Phi = 2\pi N \int v f(v) dv \int \rho d\rho \{S_2^* - 1\}_{av}. \quad (6)$$

The problem of performing the average in Eq. (6) has now been reduced to the one-state case considered explicitly by SC. This is a simplification of the more general two-state case considered by Cooper.¹²

In the one-state case (see SC) the angular average of the S matrix elements is given by

$$\begin{aligned} \{ \langle 2lm | S_2^* - 1 | 2lm \rangle \}_{av} &= (2l+1)^{-1} \\ &\times \left(\sum_{m=-l}^{+l} \langle \langle 2lm | S_{2c}^* | 2lm \rangle - 1 \rangle \right) \equiv \Gamma(l), \end{aligned} \quad (7)$$

where the subscript c on the right-hand side indicates that the interaction was evaluated in the collision axes (see Appendix).

l takes on the values zero and one. The $s(l=0)$ and $p(l=1)$ Φ matrix elements may now be written

$$\begin{aligned} \langle 200 | \Phi | 200 \rangle &= 2\pi N \int v f(v) dv \int \rho d\rho \Gamma(0), \\ \langle 21m | \Phi | 21m \rangle &= 2\pi N \int v f(v) dv \int \rho d\rho \Gamma(1). \end{aligned} \quad (8)$$

$\Gamma(0)$ and $\Gamma(1)$ [Eqs. (7)] were found, using the nonzero terms in the multipole interaction for the $n=2$ state given in the Appendix, solving numerically the relevant set of coupled Eq. (5), and substituting the resultant $\langle 2lm | S_{2c}^* | 21m \rangle$ matrix elements into Eq. (7). $\Gamma(l)$ is found to depend on the velocity as well as on the impact parameters, and to have a real and imaginary part. Figures 1 and 2 show the real part of $\Gamma(0)$ and $\Gamma(1)$, respectively, as a function of ρ . The curves are characterized by a parameter $X = \hbar/mva_0$ (cgs units). The particular curves shown are for $X = 3$ or v (a.u.) $= \frac{1}{3}$, corresponding approximately to the most probable velocity for $T = 2 \times 10^4$ °K. Shown for comparison are the values of $\Gamma(0)$ and $\Gamma(1)$ according to GKSI and SC, which are real and independent of v . Also shown are the values

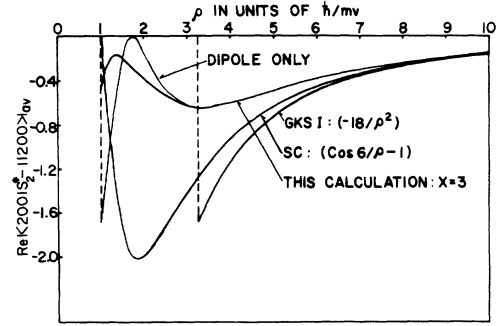


FIG. 1. The s -state matrix elements of $\{S_2^* - 1\}_{av}$ as a function of impact parameter ρ . The present calculations are a function of velocity which may conveniently be expressed through a parameter $X = \hbar/mva_0$. For the particular curve shown, $X=3$. Shown for comparison are the curves of GKSI (Ref. 1), SC (Ref. 6), and the dipole-only case. The latter three curves are independent of X .

of $\Gamma(0)$ and $\Gamma(1)$ for the "dipole only" case. In this approximation we assume that the distance (R) of the perturber electron from the origin is always greater than the distance (r) of the atomic electron and consider only the dipole interaction. This amounts to ignoring all the V^i matrix elements for $n=2$ except those for $i=1$, and setting $F(R) = -9/\sqrt{3}R$ (see Appendix). These curves are also independent of v and therefore of X . We ex-

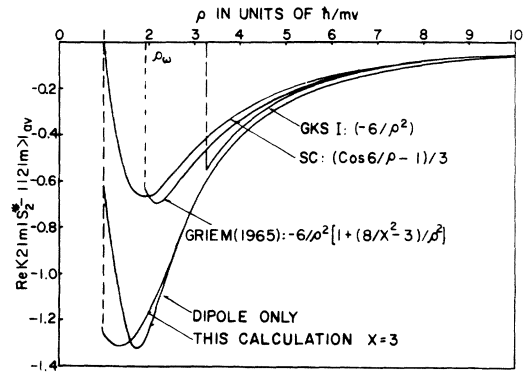


FIG. 2. The p -state matrix elements of $\{S_2^* - 1\}_{av}$ as a function of impact parameter ρ . The present calculations are a function of velocity which may conveniently be expressed through a parameter $X = \hbar/mva_0$. For the particular curve shown, $X=3$. Shown for comparison are the curves of GKSI (Ref. 1), SC (Ref. 6), and the dipole-only case. The latter three curves are independent of X . Also shown is the curve of Griem (1965) which depends on X ($X=3$ for the curve shown). The lower cutoff ρ_ω also depends on X . For $\rho < \rho_\omega$ the Lorentz-Weisskopf strong collision term is added.

pect deviations from the dipole-only case only for impact parameters less than about $\frac{1}{2}(3n^2a_0)$ (which is the radius of the state). In units of \hbar/mv this is roughly $3n^2/2X$ (or 2, for $X=3$). The deviation which occurs at approximately $2\hbar/mv$ is clearly shown in Figs. 1 and 2. The dipole-only case indicates extremely well the effect of time ordering, for without it we obtain the results of SC. For $\rho > 10\hbar/mv$ (cgs units) the $\text{Re}\Gamma(0)$ and $\text{Re}\Gamma(1)$ become equal to the GKS I values, and are independent of v . Equations (8) were integrated numerically for ρ , ranging from λ to ρ_D , and for v from $\hbar/m\rho_D$ to ∞ . The lower limit at λ corresponds roughly to the breakdown of the classical-path theory. The error in the K 's (see below) introduced by this cutoff will, because of unitarity, definitely not exceed 0.1. For ease in comparison, the resulting Φ matrix elements can be expressed as a function of electron density and temperature, by specifying the values of the constants K_1 and K_2 in the following expressions (K_1 and K_2 are themselves independent of density and depend only on the temperature):

$$\begin{aligned} \langle 200 | \Phi | 200 \rangle &= 9N(8\pi m/kT)^{1/2} \\ &\quad \times (\hbar/m)^2 [-E_1(y_{\min}) - K_1], \\ \langle 21m | \Phi | 21m \rangle &= 3N(8\pi m/kT)^{1/2} \\ &\quad \times (\hbar/m)^2 [-E_1(y_{\min}) - K_2], \end{aligned} \quad (9)$$

where $y_{\min} = (4\pi N/3m)(e\hbar A/kT)^2$,

and $E_1(y_{\min}) = \int_{y_{\min}}^{\infty} e^{-\tau} d\tau/\tau$.

Table I gives the values of K_1 and K_2 as a function of temperature, determined as indicated above. Also given for comparison are the values of K_1 and K_2 determined by SC⁶ who considered only the dipole terms in the interaction energy. We have also considered the dipole-only case. This is different from SC since it correctly takes time ordering into account. Once the impact parameter which equals $\frac{1}{2}(3n^2a_0)$ gets less than λ the dipole-only case will be a good representation of the S matrix. This occurs for $T \lesssim 5 \times 10^3$ °K. Table I clearly shows the K 's approaching the dipole-only value at low temperatures. The values of K_1 and K_2 determined by Griem,⁵ who included in an approximate way the quadrupole contribution as given by Kepple and Griem⁷ ($8kT/3E_H$), are also shown. We stress that by substituting the values of K_1 and K_2 suggested by Griem⁵ into Eqs. (9) we do not obtain the absolute values of Φ used by Kepple and Griem in their calculations because of other modifications introduced which would affect the term $-E_1(y_{\min})$. However, as mentioned in the Introduction, the effects

TABLE II. Imaginary part of the Φ matrix elements.

Temp. (°K)	$\text{Im}\langle 200 \Phi 200 \rangle/N$	$\text{Im}\langle 21m \Phi 21m \rangle/N$
5×10^3	-0.275 (-6) ^a	-0.148 (-6)
1×10^4	-0.295 (-6)	-0.128 (-6)
2×10^4	-0.317 (-6)	-0.123 (-6)
3×10^4	-0.326 (-6)	-0.132 (-6)
4×10^4	-0.332 (-6)	-0.143 (-6)
5×10^4	-0.336 (-6)	-0.154 (-6)
6×10^4	-0.339 (-6)	-0.164 (-6)

^aPower of 10 [e. g., -0.275 (-6) = -0.275×10^{-6}].

of such modifications are beyond the scope of this paper. In GKS I, $K_1=K_2=0$, and in GKS II, $K_1=K_2=1$. An upper impact cutoff at $1 \cdot \rho_D$ was used in GKS II; hence, as in the case of Griem⁵ substitution of $K_1=K_2=1$ into Eqs. (9) will not yield the absolute value of the matrix elements as used in the complete GKS II formalism.

Table II gives the imaginary parts of the Φ matrix elements as a function of electron temperature. For the electron velocities of interest the imaginary parts are essentially zero for $\rho > 10\hbar/mv$; hence the ρ integration was performed for ρ ranging from $\rho = \hbar/mv$ to $\rho = 10\hbar/mv$.

Figure 3 shows a comparison of the effects of the various estimates of K_1 and K_2 on the Lyman- α profile for an electron density of 10^{17} e/cm³ and an electron temperature of 2×10^4 °K. The profiles were calculated by substituting the various values of K_1 and K_2 given in Table I into the expression for the Φ matrix elements [Eqs. (9)], and using

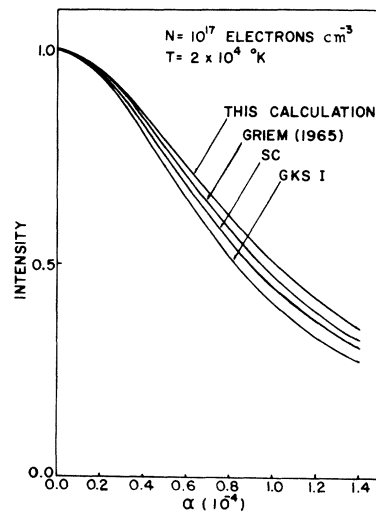


FIG. 3. Comparison of the Lyman- α profile for $N=10^{17}$ electrons/cm³ and $T=2 \times 10^4$ K according to this calculation, Griem (1965), SC (Ref. 6) and GKS I (Ref. 1).

the usual expression for the line profile [see, for example, Eq. (56) of GKSI]. The conventional reduced wavelength units α are used where $\alpha = -\Delta\lambda/F_0$ and $F_0 = 2.61 eN^{2/3}$. The profiles of Fig. 3 have been normalized to a peak intensity of one rather than to the conventional unit area. Inspection of Fig. 3 indicates that for $N = 10^{17} e/cm^3$ and $T = 2 \times 10^4$ °K the line profile calculated using the present values of K_1 and K_2 has a half-width approximately 21, 13, and 7% broader than the line profile based on estimates of K_1 and K_2 of GKSI, SC, and Griem,⁵ respectively. The GKS II-type profiles would fall between Griem⁵ and the present profile and is not shown. At temperatures below $\sim 5 \times 10^4$ °K the present profiles are found to be broader than those based on Griem⁵ while at temperatures $> 5 \times 10^4$ °K the present profiles are found to be narrower. This relative behavior of the present calculations to that of Griem⁵ should also apply to the calculations of Kepple and Griem.⁷

If we include in the line-profile calculations the imaginary parts of the Φ matrix elements given in Table II the line is found to be shifted slightly to the red. At $N = 10^{17} e/cm^3$ and $T = 2 \times 10^4$ °K the red shift is ~ 0.001 Å while at $10^{19} e/cm^3$ the shift is ~ 0.12 Å. These shifts should be independent of the upper cutoff, provided $\rho_{\max} > 10\hbar/mv$, since the imaginary part of $\Gamma(0)$ and $\Gamma(1)$ goes essentially to zero for $\rho > 10\hbar/mv$.

IV. CONCLUSION

Using the straight-line classical-path model we have calculated the impact-broadening operator Φ for the Lyman- α line. We have included proper time ordering and all orders in the multipole interaction energy (but have neglected exchange). For Lyman α this implies terms up to and including the quadrupole term (all higher terms have zero matrix elements). In general, multipoles up to $2(n-1)$ must be included, and therefore, extrapolation of the results reported here to other lines should be done with caution, if at all. A comparison between the present and previous Φ matrix elements is made in terms of two constants K_1 and K_2 . These constants differ quite considerably from the previous estimates. The numerical values of K_1 and K_2 are estimated to be accurate to ± 0.02 . K_1 is found to increase negatively with temperature, while K_2 is found to be positive and

to decrease with temperature. In contrast, K_1 and K_2 were found to be equal in previous approximations and to be independent of temperature except in the case of Griem⁵ where $K_1 = K_2$ increases positively with temperature. An average of K (Table I) is found to decrease with temperature in contrast to Griem,⁵ where the quadrupole contribution increased K . It thus seems that Kepple and Griem's⁷ estimated accuracy of $\pm 50\%$ for the quadrupole term is in error. This variation of K_1 and K_2 is significant as far as the half-width is concerned, and accounts for the fact that for a given electron density the present Lyman- α half-widths tend to be larger than those predicted using Griem⁵ for temperatures $< 5 \times 10^4$ °K and are narrower at higher temperatures. This behavior also indicates that K_2 is the significant term in determining differences between the present and previous calculations. That K_2 dominates is not entirely unexpected since it is K_2 which applies to the intense unshifted central component. Comparison between the values of K_1 and K_2 determined by SC and the values determined by the dipole-only case indicate the importance of correct time ordering when considering strong collisions (ρ small). The comparison is made in relative terms since we have used an upper cutoff of ρ_D . Provided the significant perturber velocities (e.g., v most probable v_{rms} , v_{av}) are such that $\rho_{\max} > 10\hbar/mv$ [see Eqs. (1) and (2)], the values of K_1 and K_2 should not be affected by the choice of ρ_{\max} . Similarly, the relative effects on the profiles should remain as demonstrated and be independent of the upper cutoff.

Inclusion of the imaginary part of the Φ matrix elements results in a small red shift of the line ranging from ~ 0.001 Å at 10^{17} electrons/cm³ to 0.12 Å at 10^{19} electrons/cm³. We note that as far as the shift and asymmetry of the line is concerned other effects, such as the quadratic Stark effect (see, for example, Ref. 5), might be larger than the effects we have considered.

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APPENDIX

For convenience we chose the collision axes (Fig. 4) as the coordinate system in which to evaluate the classical straight-line-path interaction of a free electron and radiating atom. The z axis is chosen to lie along the direction of \vec{p} and the x axis to lie along the direction \vec{v} . The zero of time is taken to be the time of closest approach. The origin of the coordinate system is located at the c. m. of the atomic system which, for all practical purposes, is at the nucleus. The atomic electron is located at (r, θ', ϕ') and the free electron at $(R, \theta, 0)$. Denoting the angle between R and r by β' we may write the interaction energy as

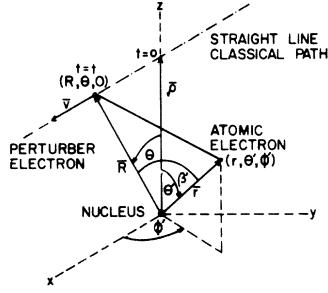


FIG. 4. Collision axes.

$$V(R) = (1/r - 1/R) + \sum_{i=1}^{\infty} (R^i/r^{i+1})P_i(\cos\beta'), \quad r > R ;$$

$$V(R) = \sum_{i=1}^{\infty} (r^i/R^{i+1})P_i(\cos\beta'), \quad r < R .$$
(A1)

The first term in the upper expression is the monopole contribution, and the index i in the summation specifies the i -pole contribution.

Using the addition theorem for spherical harmonics (the definitions given by Edmonds¹³ are used throughout) the i -th term in the interaction energy ($i \neq 0$) may be written

$$V^i(R) = 4\pi(2i+1)^{-1} \sum_{m=-i}^{+i} Y_{im}^*(\theta', \phi') Y_{im}(\theta, \phi) R^i/r^{i+1}, \quad r > R ;$$

$$V^i(R) = 4\pi(2i+1)^{-1} \sum_{m=-i}^{+i} Y_{im}^*(\theta', \phi') Y_{im}(\theta, \phi) r^i/R^{i+1}, \quad r < R .$$
(A2)

The $i=0$ term remains as in Eq. (A1).

The angular average can best be performed when the $|nlm\rangle$ wave functions are used as bases (see Cooper¹²). For this reason we choose to evaluate Eq. (3) in terms of the $|nlm\rangle$ states [Eq. (5)]. In Eq. (5) we require the matrix elements $\langle nlm | V^i | nl' m' \rangle$. Using Eqs. (A2) we get

$$\langle nlm | V^i | nl' m' \rangle = \left(\int_0^R R_{nl}^* \frac{r^i}{R^{i+1}} R_{nl'}(r) dr + \int_R^\infty R_{nl}^*(r) \frac{R^i}{r^{i+1}} R_{nl'}(r) dr \right)$$

$$\times \frac{4\pi}{(2i+1)} \int_0^{2\pi} \sin\theta' d\theta' \int_0^\pi Y_{lm}^*(\theta', \phi') Y_{l'm'}(\theta', \phi') \sum_{m''=-i}^{+i} Y_{im''}^*(\theta', \phi') Y_{im''}(\theta, \phi) d\phi' .$$
(A3)

The angular part of this expression consists of a sum of terms of the form

$$\left[\int_0^{2\pi} \sin\theta' d\theta' \int_0^\pi Y_{l_1 m_1}(\theta', \phi') Y_{l_2 m_2}(\theta', \phi') Y_{l_3 m_3}(\theta', \phi') d\phi' \right]$$

$$= \left(\frac{(2l_1+1)(2l_2+1)(2l_3+1)}{4\pi} \right)^{1/2} \begin{pmatrix} l_1 & l_2 & l_3 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_1 & l_2 & l_3 \\ m_1 & m_2 & m_3 \end{pmatrix} ,$$
(A4)

multiplied by a term $(-1)^{m_3+m_1} Y_{l_3-m_3}(\theta, \phi)$.

From the properties of the 3- j symbols [last two terms in Eq. (A4)] the l 's and m 's must satisfy the conditions

$$\vec{l}_1 + \vec{l}_2 = \vec{l}_3, \quad l_1 + l_2 + l_3 \text{ even}; \quad \text{and} \quad m_1 + m_2 = -m_3; \quad |m_1| \leq l_1, |m_2| \leq l_2, |m_3| \leq l_3 ,$$

in order for nonzero terms to exist. These conditions limit the number of nonzero terms in the interaction energy to a finite number. For a particular set of levels with principal quantum number n , $i \leq 2(n-1)$ [C. R. Vidal (private communication)].

In the case of the Lyman- α line, terms up to and including the quadrupole term contribute to the develop-

ment of the upper states ($n=2$) while only the monopole term contributes to the lower states $n'=1$. The V matrix is symmetric, and the relevant nonzero terms for the $n'=1$ and $n=2$ levels using Eq. (A3) are given below. [$n'=1$ (monopole only)]:

$$\langle 100 | V^0 | 100 \rangle = -e^{-2R}(1 + 1/R).$$

[$n=2$ (monopole, dipole, quadrupole)]:

$$\langle 200 | V^0 | 200 \rangle = -\frac{1}{8}e^{-R}(8/R + 6 + 2R + R^2),$$

$$\langle 21 \pm 1 | V^0 | 21 \pm 1 \rangle = \langle 210 | V^0 | 210 \rangle = -\frac{1}{4}e^{-R}(4/R + 3 + R + \frac{1}{6}R^2),$$

$$\langle 200 | V^1 | 210 \rangle = \rho F(R)/\sqrt{3}R,$$

$$\langle 200 | V^1 | 211 \rangle = -\langle 200 | V^1 | 21 - 1 \rangle = -vtF(R)/\sqrt{6}R,$$

$$\langle 211 | V^2 | 21 - 1 \rangle = -3(vt)^2G(R)/10R^2,$$

$$\langle 210 | V^2 | 211 \rangle = -\langle 210 | V^2 | 21 - 1 \rangle = -3\sqrt{2}vt\rho G(R)/10R^2,$$

$$\langle 210 | V^2 | 210 \rangle = (2\rho^2 - v^2t^2)G(R)/5R^2,$$

$$\langle 211 | V^2 | 211 \rangle = \langle 21 - 1 | V^2 | 21 - 1 \rangle = (v^2t^2 - 2\rho^2)G(R)/10R^2,$$

$$\text{where } F(R) = -9(1 - e^{-R} \sum_{n=0}^4 R^n/n!) / \sqrt{3} R^2,$$

$$G(R) = 30(1 - e^{-R} \sum_{n=0}^5 R^n/n!) R^3 + \frac{1}{24} R^2 e^{-R},$$

$$\text{and } R = |\vec{\rho} + \vec{v}t|.$$

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