Ionization of Lithium by Fast Protons and Electrons*

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Born's approximation is used to calculate the cross sections for ionization of lithium by fast protons (<1 Mev) and electrons (<1 kev). The electron impact results are in good agreement with those obtained by Seaton's method from experimental photoionization data. The maximum cross section for proton impact is $1.1\pi a_0^2$ at 20 kev.

INTRODUCTION

THE ionization of atomic systems by fast-proton impact is a subject of considerable interest in auroral studies and is relevant to the operation of certain types of thermonuclear devices. While there have been extensive experimental and theoretical studies of ionization by electron impact[1] (the theoretical work normally being based on Born's approximation), the agreement is poor until electron energies of the order of ten times ionization threshold are reached, even in the case of the monatomic noble gases. For these the discrepancy is almost certainly due to the failure of Born's approximation at low energies. With other gases, analysis of the data is complicated by their polyatomic nature and the inability of the present theoretical formalism readily to take account of this. Recent studies by Fite and his colleagues on monatomic oxygen[2] and hydrogen[3] indicate a similar failure of Born's approximation at low energies even for atomic hydrogen and suggest that it may persist even at moderately high energies. For atomic oxygen the comparison is made with Seaton's semiempirical approximation,[4] based on Bethe's approximation, and while the agreement is satisfactory, it yields no information on the range of validity of Born's approximation. The evidence on ionization by proton impact is less clear. Little experimental information is available at energies where Born's approximation might be expected to be valid. Theoretical studies using the Born approximation have been made by Bates and Griffing[5] for H, while He has been studied by Erskine[6] and Mapleton.[7] Erskine's work actually referred to ionization by alpha particles, but it can readily be converted to apply to electron impact and allows comparison with the experimental data of Smith[8]: the agreement is remarkably good. The improvement over earlier calculations is apparently due to replacing the p-wave part of the ejected electron wave function by that of an electron in the static field of He($1\,^2S_{\frac{1}{2}}$). The ionization of Ne by proton impact has been considered by Bates, McDowell and Omholt,[9] but only the $l=2$ part (d wave) of the ejected electron function was used, no comparison with experiment being made. An extensive experimental program of measurements of proton ionization cross sections is now in progress at the Georgia Institute of Technology at proton energies of 0.15–1.1 Mev. Results on H_2 show excellent agreement with the Born approximation calculation of Bates and Griffing,[5] provided a suitable adjustment is made to take account of the molecular structure.[10]

The present paper is intended as a beginning of a program to provide comparison data for the Georgia Tech experimental work. We are interested in the range of validity of Born's approximation, in the sensitivity of the calculations to choice of wave function, and in developing higher approximations. It is already clear from a comparison of the work of Mapleton[7] and of Dalgarno and McDowell[11] on simultaneous excitation and ionization of He by protons that such cross sections are extremely sensitive to choice of free-electron wave functions. Further difficulties arise in ensuring orthogonalization of initial and final wave functions, since if one employs single-electron atomic orbitals, those of the target and those of the residual system belong to different complete sets. With atoms of higher atomic number than He, comparison of experimental and theo-

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[6] G. A. Erskine, Proc. Roy. Soc. (London) **A224**, 362 (1954).

[7] R. A. Mapleton, Phys. Rev. **109**, 1166 (1958).

[8] P. T. Smith, Phys. Rev. **36**, 1293 (1930).

[9] D. R. Bates, M. R. C. McDowell, and A. Omholt, J. Atmospheric and Terrest. Phys. **10**, 51 (1957).

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[11] A. Dalgarno and M. R. C. McDowell in *The Airglow and Aurora*, edited by E. B. Armstrong and A. D. Dalgarno (Pergamon Press, New York, 1957), p. 340.

retical data must allow not only for the possibility of simultaneous excitation and ionization, but also for autoionization and inner-shell ionization.[12] In this paper we restrict ourselves to ionization of lithium, but consider both proton and electron impact. We employ Born's approximation and represent the ejected electron by a Coulomb function, with effective nuclear charge z. For the bound electrons we use the atomic orbitals given by Wu and Yu.[13] Since there does not appear to be any experimental data, we compare our results for electron impact with those obtained from Seaton's semiempirical formulas, via the experimental values for He and for photoionization of He and Li at threshold. No more than moderate agreement is to be expected, since Bethe's approximation, on which Seaton's result is based, is only valid at high energies.

THEORY

We consider a structureless particle of momentum $\mathbf{k}_i=\mu\mathbf{v}_i/h$ (in the center-of-mass system) incident on a lithium atom in its ground state. It is scattered through direction $(\theta,0)$, ionizing the outer $2s$ electron of the target atom, this electron being ejected with momentum $\mathbf{t}$ into the solid angle $d\omega$ about the directions (χ,ψ), and the incident particle being left with momentum $\mathbf{k}_f$.

The Born approximation to the cross section for this process may be expressed as[14]

$$Q(E_i)=2\pi\left(\frac{\mu}{2\pi\hbar^2}\right)^2\int_{-1}^{+1}\int_0^{t_{\max}}\int_\omega|\mathcal{J}(2s\rightarrow c)|^2 \times d\omega dt d\cos\theta, \quad (1)$$

where μ is the reduced mass of the colliding systems, and $\mathcal{J}(2s\rightarrow c)$ is the relevant matrix element. Transforming to momentum space, and defining

$$\mathbf{K}=\mathbf{k}_i-\mathbf{k}_f, \quad (2)$$

we can write this as

$$Q(E_i)=\frac{8}{\mathcal{S}_i^2}\int_{K_{\min}}^{K_{\max}}\int_0^{t_{\max}}\int_\omega|L(2s\rightarrow c)|^2 \times d\omega dt\, K^{-3}dK(\pi a_0^2), \quad (3)$$

in which

$$\mathcal{S}_i^2=\tfrac{1}{2}mv_i^2/I_{\mathrm{H}} \quad (4)$$

is the energy (in rydbergs) the incident particle would have if its mass were that of an electron, and

$$L(2s\rightarrow c)=\int\phi(2s,r)e^{i\mathbf{K}\cdot\mathbf{r}}\psi_t^{-*}(\mathbf{r},z)d\mathbf{r}. \quad (5)$$

Here $\mathbf{r}$ is the position vector of the ejected electron, $\phi(2s,\mathbf{r})$ is a $2s$-orbital of lithium, and $\psi_t^-(\mathbf{r},z)$ is a Coulomb wave function describing an outgoing electron in the field of a positive charge ze. We have assumed in obtaining (5) that (i) the $1s$ orbital of Li^+ is orthogonal to that of Li, and (ii) $\psi_t^-(\mathbf{r},z)$ is orthogonal to the $1s$ and $2s$ orbitals of lithium. Following Wu and Yu,[13] we adopt the trial functions

$$\psi(1s,\mathbf{r})=\frac{\alpha^{\frac{3}{2}}}{\pi^{\frac{1}{2}}}e^{-\alpha r}, \qquad \alpha=2.694, \quad (6)$$

$$\phi(2s,\mathbf{r})=N(2s)\{1-\tfrac{1}{2}(\alpha+\beta)r\}e^{-\beta r}, \quad \beta=0.764, \quad (7)$$

for the ground state of lithium, α and β being variationally determined parameters, and ψ and ϕ being orthogonal. An examination of the cross sections for the processes

$$\mathrm{Li}(1s^22s)+e\rightarrow\mathrm{Li}(1s2s2p,\ \text{or}\ 1s^22p,\ \text{or}\ 1s2p^2)+e \quad (8)$$

by one of us (G.P.) with various forms of variational functions for the orbitals reveals that these cross sections are not very sensitive to slight improvements in the wave functions.[15]

Our choice of orthogonalization requires ψ_t^- to be orthogonal to $\psi(1s)$ and $\phi(2s)$. By taking $z=\alpha$, we can satisfy the first requirement but not the second. Physically, the ejected electron will be well screened by the inner electrons, and it might be supposed that $z=\beta$ or $z=1.0$ would be a better choice. However, with these values of z neither orthogonalization condition is satisfied. It should in principle be possible to form a linear combination of $\psi_t^-(\beta,\mathbf{r})$, $\psi(1s,\alpha)$, and $\phi(2s,\beta)$ which satisfies both conditions and retains the correct asymptotic properties, but the algebra would be so complex that it would be just as simple to perform a Hartree-Fock solution for the ejected electron. We hope to do this at a later date. For the present we adopt $z=\alpha$ to obtain the greatest simplification of the analysis. The resultant wave function at least has the correct properties for small r, from which the dominant contribution to the matrix element arises.[14,16] Writing then

$$\psi_t^-(\mathbf{r},z)=\frac{t}{(2\pi)^{\frac{3}{2}}}e^{\pi z/2t}\Gamma\left(1-\frac{iz}{t}\right) \times\exp(i\mathbf{t}\cdot\mathbf{r})F\left(\frac{iz}{t},1,-itr-i\mathbf{t}\cdot\mathbf{r}\right), \quad (9)$$

where we have normalized so that

$$\int d\omega\int\psi_t^{-*}\psi_{t'}^-d\mathbf{r}=\delta(\mathbf{t}'-\mathbf{t}), \quad (9a)$$

[12] A. M. Arthurs and B. L. Moiseiwitsch, Proc. Roy. Soc. (London) **A247**, 550 (1958).
[13] T. Y. Wu and F. C. Yu, Chinese J. Phys. **5**, 162 (1944).
[14] N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Oxford University Press, New York, 1949), 2nd ed. Chap. XI.
[15] G. Peach, Ph.D. thesis, London, 1960 (unpublished).
[16] E. H. S. Burhop, Proc. Cambridge Phil. Soc. **36**, 43 (1940).

straightforward analysis[3,17] yields

$$Q(E_i)=\frac{2^8\beta^5 z}{5\epsilon}S_i^{-2}\int_0^{t_{\max}}\int_{K_{\min}}^{K_{\max}}F(t,K)\times\exp\left\{-\frac{2z}{t}\arctan\frac{2\beta t}{\beta^2+K^2-t^2}\right\}\times(1-e^{-2\pi z/t})^{-1}(c^2+d^2)^{-4}(a^2-b^2)^{-5}\times t^{-3}K^{-3}dtdK\ (\pi a_0^2),\quad(10)$$

where

$$a=\beta^2+K^2+t^2,\quad b=2Kt,\quad c=\beta^2+K^2-t^2,\quad d=2\beta t,\quad \epsilon=\alpha^2+\alpha\beta+\beta^2,\quad(11)$$

and $F(t,K)$ is a complicated polynomial in β, t, K given in the Appendix.

The most important contribution to the cross section, at energies much above threshold, should be the contribution from transitions to the p wave of the continuum. To obtain this we replace (9) by its p wave,

$$\psi_i^-(z;\,p\text{ wave})=\frac{i\cos\delta}{6\pi}\left[\frac{tz(t^2+z^2)}{1-e^{-2\pi z/t}}\right]^{\frac{1}{2}}re^{itr}F\left(2+\frac{iz}{t},\,4,\,2itr\right),\quad(12)$$

retaining the normalization (9a) and putting $\hat{t}\cdot\hat{r}=\cos\delta$. Evaluating (5) using (12), we adopt an analysis developed by Bates *et al.*[9] and obtain

$$Q_p(E_i)=\frac{18\beta^5 z}{S_i^2}\int_{K_{\min}}^{K_{\max}}\int_0^{t_{\max}}\frac{(t^2+z^2)}{t^7K^7}(1-e^{-2\pi z/t})^{-1}\times\exp\left\{-\frac{2z}{t}\arctan\frac{2\beta t}{\beta^2+K^2-t^2}\right\}\times\{(\phi_1+\psi_2)\cos\Theta+(\phi_2-\psi_1)\sin\Theta\}^2dKdt(\pi a_0^2),\quad(13)$$

in which

$$\Theta=\frac{z}{t}\log_e\left|\frac{\beta-it-iK}{\beta+it-iK}\right|,\quad(14)$$

and

$$\begin{aligned}
\phi_1&=8Kt^4(t^2+z^2)^{-1}(c^2+d^2)^{-1}[(\beta-z)c+2\beta(t^2+\beta z)],\\
\psi_1&\equiv 4t^2(t^2+z^2)^{-1}\{a(c^2+d^2)^{-1}\times[c(t^2+\beta z)-2\beta t^2(\beta-z)]-\beta z^{-1}(t^2+z^2)\},\\
\phi_2&\equiv 2\delta'\{\beta(c^2+d^2)^{-2}(c^2-d^2+4ct^2)(a^2+b^2)\\
&\quad-2a(t^2+z^2)^{-1}(c^2+d^2)^{-1}(2\beta ct^2+zct^2+2\beta z^2c+2\beta z^2t^2)\\
&\quad+z^{-1}(3\beta z+2t^2)\},\\
\psi_2&\equiv 8t^2K\delta'(c^2+d^2)^{-2}\{a(c^2-d^2-4\beta^2c)\\
&\quad-(t+z)^{-1}(a^2-b^2)(z^2c-d^2-2\beta zt^2-4\beta z^2)\},\quad(15)\\
\delta'&=\tfrac{1}{3}(\alpha+\beta).
\end{aligned}$$

[17] H. S. W. Massey and C. B. O. Mohr, Proc. Roy. Soc. (London) **A140**, 613 (1933).

Considerable cancellation occurs for small K; a power series expansion of $\{(\phi_1+\psi_2)\cos\Theta+(\phi_2-\psi_1)\sin\Theta\}$ was therefore developed, the leading term being K^{+3}. This implies that at large E_i the p-wave contribution to the cross section decreases as $E_i^{-1}\log_e E_i$, as it should.

As a further check on the accuracy of the two formulations we made use of Seaton's method[4] of deriving the cross section for ionization by electron impact Q_X of a species X from known results for a species Y together with a knowledge of the photoionization cross section at threshold for both X and Y. This has been applied successfully to electrons on N and O (using results on Ne) by Seaton.[4] It is essential[9] to choose as comparison species one whose outer electrons have the same orbital angular momentum. We used Smith,[8] Erskine,[6] and Massey's[17] results on ionization of helium together with the values[18–20] (which are further discussed elsewhere[15]),

$$a_{\rm Li}(0)=3.70\times10^{-18}\text{ cm}^2,\quad a_{\rm He}(0)=7.80\times10^{-18}\text{ cm}^2\quad(16)$$

for the relevant photoionization cross sections. Then

$$I_AQ_A(I_A\epsilon)/a_A(0)\approx I_BQ_B(I_B\epsilon)/a_B(0),\quad(17)$$

where I_A is the ionization potential of species A, and the cross sections are compared at ϵ units above threshold.

DISCUSSION

Ionization cross sections were readily computed from the above results. We have

$$K_{\min}=k_i-k_f,\quad K_{\max}=k_i+k_f,\quad(18)$$

and $t_{\max}$ is given by

$$\frac{k_i^2}{2\mu}-\epsilon_i=\frac{k_f^2}{2\mu}+\frac{t^2}{2m_e}.\quad(19)$$

For electron impact we use Eq. (18) but note that for proton impact $K_{\max}$ and $t_{\max}$ may be taken as infinite; and

$$K_{\min}\simeq\frac{t^2+\epsilon_i}{2S_i}\left\{1+\frac{2m_e}{7M_pS_i^2}(\epsilon_i+t^2)+\cdots\right\},\quad(20)$$

where ϵ_i is the (positive) binding energy of the initial state.

The electron impact results are shown in Fig. 1 as computed from Eqs. (13) and (17). The p-wave results are well approximated by Eq. (17) except near threshold, where (17), based as it is on an expansion in powers of K, is not expected to be reliable, and where the contribution from the s wave might be expected to be most important. It is unlikely that higher partial waves make a significant contribution in this case. The value of $a_{\rm Li}(0)$ chosen is open to some criticism. More recent

[18] D. R. Bates, Monthly Notices Roy. Astron. Soc. **106**, 423, 432 (1946).
[19] J. A. Wheeler, Phys. Rev. **43**, 258 (1933).
[20] R. W. Ditchburn, Proc. Roy. Soc. (London) **A236**, 216 (1956).

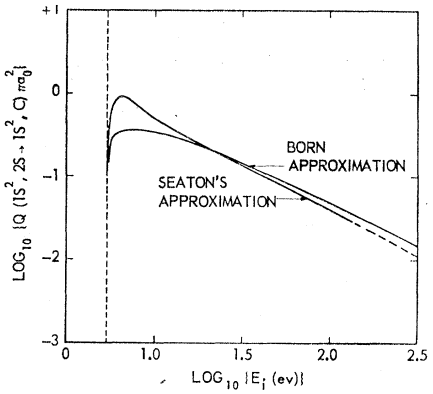

FIG. 1. Ionization of lithium by electron impact. Seaton's approximation and Born approximation (p wave, $\alpha=z$).

experimental work by Tunstead[21] suggests $a_{\mathrm{Li}}(0)=2.5\times10^{-18}$ cm^2, while a calculation by Stewart[22] using a Hartree-Fock p wave yields $a_{\mathrm{Li}}(0)=1.2\times10^{-18}$ cm^2. Adoption of Tunstead's value would reduce the value given by about 50%. Nevertheless an examination of Q_{ioniz} (p wave) as a function of z makes it clear that $z=\alpha$ still gives the best agreement. We have used the wave function ψ_t^- (z, p wave) given by Eq. (12) to calculate $a_{\mathrm{Li}}(0)$ (making use of the formulas presented by Bates[18]), as a function of z. We find that it varies as $z^7\exp(-bz)$, where b is a constant. It is then possible to pick z so that $a_{\mathrm{Li}}(0,z)$ is equal to the experimental value. This yields $z=1.29$, and gives an ionization cross section about a factor of ten larger than that predicted by the Seaton method for the same value of $a_{\mathrm{Li}}(0)$. However, when we examine the proton-impact cross sections (Fig. 2), while it is gratifying to have close agreement between results calculated from (10) and (13) below and close to the maximum, Born's approximation cannot be regarded as satisfactory at these energies. The p-wave cross sections show the expected dependence on impact energy and should be

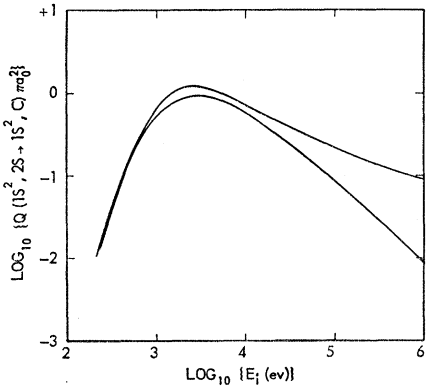

FIG. 2. Ionization of lithium by proton impact. Lower curve, p-wave Born approximation, $\alpha=z$; upper curve, Eq. (10), $\alpha=z$.

[21] J. Tunstead, Proc. Phys. Soc. (London) **A66**, 304 (1953).
[22] A. L. Stewart, Proc. Phys. Soc. (London) **A67**, 917 (1954).

reliable at high energies. The reason for the failure of Eq. (10) at high energies is of course the lack of orthogonality of $\phi(2s)$ and $\psi_t^-(\alpha,\mathbf{r})$. An expansion of $e^{i\mathbf{K}\cdot\mathbf{r}}$ in powers of K, for small K, shows that $|L(2s\rightarrow c)|^2$ is not vanishing at high energies. It is not quite independent of energy since it remains a function of t. The p-wave (lower) curve in Fig. 2 is probably the better estimate of the cross section.

If complete screening is assumed we may take $z=1$ and readily compute the p-wave cross section, since the p-wave part of ψ_t^- is automatically orthogonal to all the bound-state orbitals employed, and Eq. (13) is valid for all z. For proton impact the maximum cross section occurs at the same energy but its absolute value is increased by a factor of three while at 1 Mev, it is larger by a factor of seven. The change in the electron impact cross sections is very similar: at 100 ev the p-wave cross section for $z=1$ exceeds that for $z=\alpha$ by a factor of 6.4.

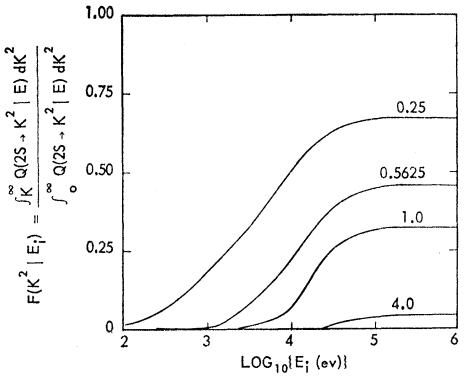

FIG. 3. Ionization of lithium by proton impact. Fraction of electrons ejected with energy greater than K^2I_{H} (numbers on curves give value of K^2).

We have repeated the calculations of Wu and Yu[13] on double excitation and are in substantial agreement with them.[15] For electron impact the most important of these processes is

$$\mathrm{Li}(1s^22s)+e\rightarrow\mathrm{Li}(1s2s2p\,{}^2P)+e, \qquad (21)$$

which has a maximum cross section of $2\times10^{-2}\pi a_0^2$ at 150 ev and falls off rapidly with increasing impact energy. It is clear that autoionization cannot play a significant role in impact ionization of lithium. The contributions from simultaneous excitation and ionization have been computed by Wu and Yu.[13] However, their use of a plane wave to represent the ejected electron is unsatisfactory and their results are of uncertain accuracy. Even if they were larger by an order of magnitude they would nowhere exceed 20% of our p-wave contribution. Studies of this process in helium[11,12] show that cross sections for simultaneous excitation and ionization never exceed 10% of those for simple outer-electron ionization, whatever assumptions as to orthogonality are adopted. Inner shell ionization will also

contribute; from the data presented by Arthurs and Moisiewitsch,[12] who show that at a given ratio of E_i to ionization potential the K-shell ionization cross section is proportional to z^2, we estimate that the cross section for this process will not exceed 4.5×10^{-20} cm^2 in lithium.

For the sake of its interest in studies of secondary ionization and auroral processes we display in Fig. 3 the fraction of the number of ejected electrons (p wave) having energy in excess of K^2I_H, as a function of impact energy. At high energies more than 30% of these electrons have energy greater than I_H, in close agreement with the results of Bates and Griffing[5] for $1s$ electrons, but only about half as many as for $2p$ electrons.[9]

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APPENDIX

The function $F(t,K)$ introduced in Eq. (10) is given by

$$\begin{aligned}F(t,K)=&3(5a^4+10a^2b^2+b^4)(f_1^2+f_4^2)\\&+12ab(5a^2+3b^2)(f_1f_2+f_4f_5)\\&+(5a^4+38a^2b^2+5b^4)(f_2^2+2f_1f_3+2f_4f_6+f_5^2)\\&+12ab(5b^2+3a^2)(f_2f_3+f_5f_6)\\&+3(5b^4+10a^2b^2+a^4)(f_3^2+f_6^2),\end{aligned}\tag{A-I}$$

in which

$$\begin{aligned}&f_1=at(c^2+d^2)g_1-\delta g_5, \quad f_2=t(ag_2-bg_1)(c^2+d^2)-\delta g_6,\\&f_3=-tb(c^2+d^2)g_2-\delta g_7, \quad f_4=at(c^2+d^2)g_3-\delta g_8,\\&f_5=t(ag_4-bg_3)-\delta g_9, \quad f_6=-tb(c^2+d^2)g_4-\delta g_{10},\end{aligned}\tag{A-II}$$

where

$$\begin{aligned}&g_1=za(tc-\beta d)+\beta t(c^2+d^2), \quad g_2=zb(\beta d-tc),\\&g_3=za(\beta c+td)-z\beta(c^2+d^2), \quad g_4=-zb(\beta c+td),\\&g_5=a^2y_1+ay_2+2\beta^2(c^2+d^2)^2(2t^2-z^2),\\&g_6=-2aby_1-by_2, \quad g_7=b^2y_1,\\&g_8=a^2y_3+ay_4-6zt\beta^2(c^2+d^2)^2,\\&g_9=-2aby_3-by_4, \quad g_{10}=b^2y_3,\end{aligned}\tag{A-III}$$

and

$$\begin{aligned}y_1=&4zt\{\beta t(c^2-d^2)-cd(\beta^2-t^2)\}\\&-2z^2\{(\beta^2-t^2)(c^2-d^2)+4\beta tcd\}+ztd(c^2+d^2),\\y_2=&4z\beta(c^2+d^2)\{z(\beta c+td)-t(\beta d-tc)\}-t^2(c^2+d^2)^2,\\y_3=&4z^2\{\beta t(c^2-d^2)-cd(\beta^2-t^2)\}\\&+2zt\{(\beta^2-t^2)(c^2-d^2)+4\beta tcd\}-ztc(c^2+d^2),\\y_4=&4z\beta(c^2+d^2)\{t(\beta c+td)+z(\beta d-tc)\}\\&+tz(c^2+d^2)^2.\end{aligned}\tag{A-IV}$$