ANGULAR DISTRIBUTION IN INELASTIC ATOMIC COLLISIONS

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Formulas for the differential cross section of elastic scattering are obtained in the approximation of two interacting terms and are used to analyze the scattering of He^+ by Ne. It is shown that the slope of the potential of the inelastic interaction at the point of term intersection can be obtained from the period of the oscillations of the inelastic cross section.

1. INTRODUCTION

THE differential cross section of inelastic atomic collisions has been intensively investigated recently. The results of such investigations yield extensive information on the electronic states of a two-particle system, and this information can be used to verify the theoretical predictions. By now, measurements have already been made of the differential cross section of elastic scattering of He^+ by Ne and Ar in a wide range of energies from 10 to 600 eV[1]. Starting with 30 eV for He^+ + Ar and 50 eV for He^+ + Ne, regular oscillations have been observed on the smooth plot of the elastic-scattering cross section. In the experiment with Ne, the amplitude of the oscillations is smaller than in the experiment with Ar. In both cases, they begin with a certain value of the reduced angle $\tau \equiv \theta E$ (θ—scattering angle, E—c.m.s. energy). With increasing energy of the incident ions, the value of τ first decreases slowly, but then rapidly assumes a constant value, which is definite for each system. The quasiclassical expression $E\theta(\rho)$ can be expanded in powers of E^{-1}, and the coefficients of the series depend only on the impact parameter ρ[2]:

$$\tau \equiv \theta E = \tau_0(\rho) + E^{-1}\tau_1(\rho) + \ldots$$

The interaction causing the transition differs from zero in a small vicinity of the point of intersection of the electronic terms, and its influence on the scattering is large if the particles pass through this region sufficiently slowly, i.e., in those cases when the classical turning point coincides with the level intersection point. At large energies $\tau \approx \tau_0$ and the impact parameter coincides, with the same accuracy, with the turning point. Therefore, if the oscillations begin at τ = const, then it can be assumed that they are the result of the interaction of the electronic terms at the internuclear distance $R_0 \approx \rho$.

In this paper, using the model of linear terms, we obtain an expression for the differential cross section of inelastic scattering and attempt to estimate from the period of the oscillations, the slope of the terms for He^+ + Ne at the intersection point. It turns out here that a transition occurs from the ground state of He^+ + Ne to an excited state whose electronic term corresponds to attraction at the intersection point.

2. SCATTERING MATRIX IN THE APPROXIMATION OF TWO STATES

To calculate the transition probabilities in slow collisions of atoms and molecules, frequent use is made of the approximation of two molecular terms. The Schrödinger equation for the two-level problem is

$$\left[-\frac{\hbar^2}{2\mu}\Delta_R + \hat{H}^{\text{эл}}(\mathbf{r}, \mathbf{R})\right]\psi = E\psi, \tag{1}$$

and its solution is sought in the form

$$\psi = \varphi_1(\mathbf{r}) \sum_{l, m} \frac{1}{R}\chi_{1l}(R) Y_{lm}(\theta, \varphi) + \varphi_2(\mathbf{r}) \sum_{l, m} \frac{1}{R}\chi_{2l}(R) Y_{lm}(\theta, \varphi), \tag{2}$$

where $\varphi_1(\mathbf{r})$ and $\varphi_2(\mathbf{r})$ are the wave functions of the electrons in the atoms, μ the reduced mass of the nuclei, and $\mathbf{R}$ the radius vector of their relative position. The role of the nuclear wave functions is assumed by $\chi_{il}(R)$. They satisfy the system of two equations

$$\frac{\hbar^2}{2\mu}\chi_{1l}''(R) + \left(E - H_{11} - \frac{\hbar^2 l(l+1)}{2\mu R^2}\right)\chi_{1l}(R) = H_{12}\chi_{2l}(R),$$
$$\frac{\hbar^2}{2\mu}\chi_{2l}''(R) + \left(E - H_{22} - \frac{\hbar^2 l(l+1)}{2\mu R^2}\right)\chi_{2l}(R) = H_{21}\chi_{1l}(R). \tag{3}$$

Far from the term intersection point, the system breaks up, and the motion of the nuclei becomes quasiclassical in potentials $H_{11}(R)$ and $H_{22}(R)$.

The region of the transition is assumed to be small, and therefore $H_{ik}(R)$ and $W_l(R) = \hbar^2 l(l+1)/2\mu R^2$ can be expanded in powers of $x = R - R_0$ (R_0 is the point of intersection of the potentials $H_{11}(R)$ and $H_{22}(R)$. In the expansions of $H_{ii}(R)$ and $W_l(R)$ we retain the terms that are linear in x, and in the expansion of $H_{ik}(R)$ only the zeroth term. Then, within the region of transition, we have for the functions $\chi_{il}(R)$ the system of two equations

$$\frac{\hbar^2}{2\mu}\chi_{1l}'' + [E(l) + F_1(l)x]\chi_{1l} = a\chi_{2l},$$
$$\frac{\hbar^2}{2\mu}\chi_{2l}'' + [E(l) + F_2(l)x]\chi_{2l} = a\chi_{1l}, \tag{4}$$

where

$$E(l) = E - H_{ii}(R_0) - W_l(R_0),$$
$$F_i(l) = -\frac{\partial}{\partial R}[H_{ii} + W_l]|_{R=R_0}, \quad a = H_{12}(R_0). \tag{5}$$

The solution of the system (4) is made continuous with the quasiclassical solution outside the region of the transition, and is asymptotically continued to infinity, where the wave function of the system of the two nuclei can be written in the form of a column

$$\hat{\chi}_l = \begin{pmatrix} \alpha_1\chi_{1l}^{(-)} - \beta_1\chi_{1l}^{(+)} \\ \alpha_2\chi_{2l}^{(-)} - \beta_2\chi_{2l}^{(+)} \end{pmatrix}, \tag{6}$$

$$\chi_{il}^{(\pm)} = (v_i)^{-1/2}\exp\{\pm i(k_iR - \pi l/2)\},$$

where the first row is the wave function of the nuclei in the first channel (motion in potential $H_{11}(R)$), and the second row is the wave function of the nuclei at the second channel (motion in potential $H_{22}(R)$). The connection between the columns α and $\hat{\beta}$, describing the amplitudes of the converging and diverging waves, is determined by the S matrix

$$\hat{\beta} = \hat{S}\hat{\alpha}. \tag{7}$$

If the splitting between the adiabatic terms ($\sim a$) is small, then the system (4) can be solved by perturbation theory. The scattering matrix S^l obtained in this case is given by

$$S_{12}^l = i\sqrt{\pi}\, b^{2/3}\Phi(-\varepsilon b^{2/3})\exp[i(\Delta_l^1 + \Delta_l^2)],$$
$$S_{11,22}^l = [1 - \pi b^{4/3}/2\Phi^2(-\varepsilon b^{2/3})]\exp(2i\Delta_l^{1,2}), \tag{8}$$

where

$$\varepsilon = \frac{E(l)(F_1 - F_2)}{2a(F_1F_2)^{1/2}}; \quad b = \frac{4a}{\hbar}\sqrt{\frac{\mu a}{(F_1 - F_2)(F_1F_2)^{1/2}}}; \quad b \lesssim 1,$$

$\Phi(-\epsilon b^{2/3})$—Airy function, $\Delta_l^{1,2}$—quasiclassical scattering phases in the potentials $H_{11}(R)$ and $H_{22}(R)$.

In the case when perturbation theory is not applicable, the S matrix can be obtained for $\epsilon \gg 1$ and $-\epsilon \gg 1$, i.e., for transitions for which the level intersection point is far from the turning points. The problem is solved by the method of Pokrovskiĭ and Khalatnikov[3]. We present the results:

When $\epsilon \gg 1$ ($b \gg 1$), the S matrix takes the form

$$S_{11}^l = [(1 - e^{-2\pi\delta})e^{-2i\eta} + e^{-2\pi\delta}]\exp(2i\Delta_l^1 - i\delta - 2i\delta\ln 8\varepsilon),$$
$$S_{22}^l = [(1 - e^{-2\pi\delta})e^{+2i\eta} + e^{-2\pi\delta}]\exp(2i\Delta_l^2 + i\delta + 2i\delta\ln 8\varepsilon), \tag{9}$$
$$S_{12}^l = +i\sqrt{2}P^{1/2}(l)\sin\eta\exp[i(\Delta_l^1 + \Delta_l^2)],$$

where $P(l) = 2e^{-2\pi\delta}(1 - e^{-2\pi\delta})$ is the Landau-Zener formula for the probability of a non-adiabatic transition, $\delta = b\epsilon^{-1/2}/8$,

$$\eta = \frac{2}{3}b\varepsilon^{3/2} - \delta + \delta\ln\delta + \frac{\pi}{4} + \frac{i}{2}\ln\frac{\Gamma(1+i\delta)}{\Gamma(1-i\delta)} \tag{9'}$$
$$\approx \begin{cases} {}^2/_3 b\varepsilon^{3/2} + \pi/4 & \delta \ll 1 \\ {}^2/_3 b\varepsilon^{3/2} & \delta \gg 1 \end{cases};$$

When $-\epsilon \gg 1$ ($b \gg 1$) we get

$$S_{11}^l = \exp[2i(\Delta_l^1 + \pi\delta)]; \quad S_{22}^l = \exp[2i(\Delta_l^2 - \pi\delta)];$$
$$S_{12}^l = i\frac{\sqrt{2\pi\delta}}{\Gamma(1-\delta)}\delta^{-\delta}e^{\delta}\exp\left[-\frac{2}{3}b|\varepsilon|^{3/2} + i(\Delta_l^1 + \Delta_l^2)\right] \tag{10}$$
$$\approx \begin{cases} i\sqrt{2\pi\delta}\exp(-{}^2/_3 b|\varepsilon|^{3/2} + i(\Delta_l^1 + \Delta_l^2)) & \text{for } \delta \ll 1 \\ 2i\sin\pi\delta\exp(-{}^2/_3 b|\varepsilon|^{3/2} + i(\Delta_l^1 + \Delta_l^2)) & \text{for } \delta \gg 1 \end{cases}$$

where $\delta = (1/8)b|\epsilon|^{-1/2}$ ($|S_{12}^l|^2$ gives the probability of the nonadiabatic transition, which coincides with the results of[4]).

3. DIFFERENTIAL INELASTIC-SCATTERING CROSS SECTION

In this section, using the expressions for the S matrix, we obtain formulas for the differential inelastic-scattering cross section. The inelastic-scattering amplitude equals[5]

$$f_{12}(\theta) = \frac{1}{2i\sqrt{k_1k_2}}\sum_l (2l+1)P_l(\cos\theta)S_{12}^l. \tag{11}$$

Inasmuch as the motion of the nuclei is considered in the quasiclassical approximation, we can change in (11) from summation to integration with respect to l, and replace $P_l(\cos\theta)$ by its asymptotic form at $l \gg 1$.

We consider first the case of strong splitting of the terms. For $\epsilon \gg 1$, the off-diagonal element of S_{12}^l is given by (9). Substituting it in (11), we get

$$f_{12}(\theta) = \frac{1}{2\sqrt{\pi k_1k_2\sin\theta}}\int_0^\infty dl(l+1/2)^{1/2}P^{1/2}(l) \tag{12}$$
$$\times[e^{i(\varphi_+ + \eta)} - e^{i(\varphi_+ - \eta)} + e^{i(\varphi_- - \eta)} - e^{i(\varphi_- + \eta)}],$$

where

$$\varphi_\pm = \Delta_l^1 + \Delta_l^2 \pm \theta(l + 1/2) \pm \pi/4. \tag{13}$$

The integral with respect to l is calculated by the saddle-point method. It is assumed here that the phase η is small compared with the quasiclassical phases $\Delta_l^{1,2}$. Therefore, for potentials that are repulsion fields, only to the last two exponentials will have a saddle point. The differential inelastic-scattering cross section equals

$$\frac{d\sigma_{12}}{d\theta} = \frac{\pi}{k_1^2}\left[\frac{P(l)(l+1/2)}{|\ddot{\varphi}_- - \ddot{\eta}|}\bigg|_{l=l_\mathrm{I}} + \frac{P(l)(l+1/2)}{|\ddot{\varphi}_- + \ddot{\eta}|}\bigg|_{l=l_\mathrm{II}}\right. \tag{14}$$
$$\left. -2\left(\frac{P(l)(l+1/2)}{|\ddot{\varphi}_- - \ddot{\eta}|}\right)^{1/2}_{l=l_\mathrm{I}}\left(\frac{P(l)(l+1/2)}{|\ddot{\varphi}_- + \ddot{\eta}|}\right)^{1/2}_{l=l_\mathrm{II}}\cos(\Omega^-(l_\mathrm{I}) - \Omega^+(l_\mathrm{II}))\right],$$

where l_I and l_II are the roots of the equations

$$\frac{d}{dl}(\varphi_- \pm \eta) = 0; \quad \ddot{\varphi}_- = \frac{d^2}{dl^2}\varphi_-; \quad \ddot{\eta} = \frac{d^2\eta}{dl^2},$$

and the function in the argument of the cosine is given by

$$\Omega^\pm(l) = \varphi_- + \frac{\pi}{4} + \frac{i}{2}\ln\frac{\ddot{\varphi}_- \pm \ddot{\eta}}{|\ddot{\varphi}_- \pm \ddot{\eta}|} \pm \eta. \tag{14'}$$

Thus, $d\sigma_{12}/d\theta$ oscillates with a period $\sim 2\pi/[\Omega^-(l_\mathrm{I}) - \Omega^+(l_\mathrm{II})]$ between the envelopes

$$\frac{d\sigma_\pm}{d\theta} = \frac{\pi}{k_1^2}\left[\left(\frac{P(l)(l+1/2)}{|\ddot{\varphi}_- - \ddot{\eta}|}\right)^{1/2}_{l=l_\mathrm{I}} \pm \left(\frac{P(l)(l+1/2)}{|\ddot{\varphi}_- + \ddot{\eta}|}\right)^{1/2}_{l=l_\mathrm{II}}\right]^2. \tag{15}$$

For an under the barrier transition ($-\epsilon \gg 1$), oscillations in the inelastic cross section will occur when $\delta \gg 1$ (see formula (10)). In this case, the expression for $d\sigma_{12}/d\theta$ is similar to (14), where the phase η will be replaced by $\pi\delta$ and $P(l)$ will be replaced by the tunnel-transition probability $P_T(l) = 2\exp[-(4/3)b|\epsilon|^{3/2}]$.

For small splitting of the adiabatic terms the off-diagonal element of the S matrix is given by (8). This formula was obtained for all values of ϵ, i.e., for all values of the kinetic energy at the intersection point.

The amplitude of inelastic scattering in a repulsion field is

$$f_{12}(\theta)=\frac{i}{2\sqrt{2\pi k_1k_2\sin\theta}}\int\limits_{-\infty}^{+\infty}du e^{iu^3/3}\int\limits_0^{\infty}dl(l+{}^1/_2)^{1/2}b^{2/3}(l)$$

$$\times\exp\left\{i\left[\Delta_l^1+\Delta_l^2-\theta(l+{}^1/_2)-\frac{\pi}{4}-u\varepsilon b^{2/3}\right]\right\}. \quad (16)$$

We have used here the definition of the Airy function

$$\Phi(-x)=\frac{1}{\sqrt{\pi}}\int\limits_0^{\infty}\cos\left(\frac{u^3}{3}-ux\right)du. \quad (17)$$

We expand the integrand in (16) in powers of $(l - l_0)$, where l_0 is the threshold value of the orbital angular momentum, defined by the condition

$$E(l_0)=E-H_{ii}(R_0)-\hbar^2(l_0+{}^1/_2)^2/2\mu R_0^2=0. \quad (18)$$

We retain in the exponential the terms $\sim(l - l_0)^2$, and take the pre-exponential function at the point l_0 (the parameter of the expansion is $(l - l_0/l_0$, which depends on the second integration variable u. The possibility of such an expansion will be justified in what follows.)

After calculating the integral with respect to l, we obtain an expression for the differential inelastic-scattering cross section in the form

$$\frac{d\sigma_{12}}{d\theta}=\frac{2\pi(l_0+{}^1/_2)}{k_1^2}\frac{b^{4/3}(l_0)}{4d^2(\varepsilon b^{2/3})dl_0^2}I(\theta-\theta_0), \quad (19)$$

where

$$I(\theta-\theta_0)=\left|\int\limits_{-\infty}^{+\infty}(\lambda+u)^{-1/2}\exp\left\{i\left[\frac{u^3}{3}+\frac{(\gamma(\theta-\theta_0)-\nu u)^2}{\lambda+u}\right]\right\}du\right|^2. \quad (20)$$

The parameters which enter in the integral are as follows: $\theta_0 = d(\Delta_l^1 + \Delta_l^2)/dl_0$—threshold scattering angle[1],

$$\lambda=\frac{1}{2}|\dot{\theta}_0|\left(\frac{\hbar^2}{2\mu R_0^2}\right)^{-2/3}(R_0F_1)^{2/3}\left(\frac{1-x}{x}\right)^{-2/3}Q^{-1}, \quad (21)$$

$$\gamma=\frac{1}{2}\left(\frac{\hbar^2}{2\mu R_0^2}\right)^{-1/6}(R_0F_1)^{1/3}\left(\frac{1-x}{x}\right)^{-1/6}Q^{-1/2},$$

$$\nu=\left(\frac{\hbar^2}{2\mu R_0^2}\right)^{-1/6}(R_0F_1)^{-1/3}\left(\frac{1-x}{x}\right)^{1/3}\sqrt{E-H(R_0)}Q^{-1/2},$$

where

$$x=\frac{F_2}{F_1},\quad \dot{\theta}_0=\frac{d^2}{dl_0^2}(\Delta_l^1+\Delta_l^2),\quad Q=\frac{16}{3}\frac{E-H(R_0)}{R_0F_1}\frac{1+x}{x}-1. \quad (22)$$

A plot of the differential cross section, calculated in accordance with formula (19) for certain values of the parameters (see Sec. 4) is shown in Fig. 1. The oscillations in the cross section, just as in the case of large splitting of the adiabatic terms, are due to the interference of the amplitudes corresponding to scattering by two different potentials.

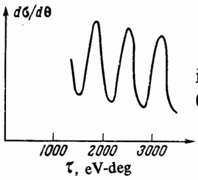


FIG. 1. Differential cross section of scattering of He^+ by Ne at E_{lab} = 400 eV and F_2/F_1 = 0.14. The absolute scale is arbitrary.

[1]If the position of the threshold θ_0 decreases with increasing energy, then $d\theta_0/dE < 0$, from which it follows that $d\theta_0/dl_0 = \dot{\theta}_0 < 0$.

4. ANALYSIS OF SCATTERING OF He^+ BY Ne

In experiments on scattering of He^+ by Ne, there are observed regular oscillations of the inelastic differential cross section. They begin with $\tau \sim 1000$ eV-deg. The minimum amplitude of the oscillations differs from zero. With increasing energy of the He^+ ions, the period of the oscillations and the minimum amplitude decrease[2]. When τ is in the interval from zero to 4000 eV-deg., the inelastic losses are small, so that it can be assumed that in this region of energies and angles only one inelastic channel is open, and the interaction between the adiabatic terms is weak. Then, using formula (19) for the differential inelastic-scattering cross section, we can obtain certain information concerning the interaction potential between the particles in the inelastic channel.

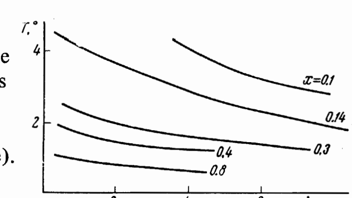
From an analysis of the data on the elastic scattering of He^+ by Ne, the following inelastic-interaction potential was obtained in[6]

$$H_{11}(R)=\frac{\xi e^2}{R}\exp\left(-\frac{R}{c}\right)$$

with parameters ξ = 17.5 and c = 0.68a, and the position of the intersection point R_0 = 8.9a was obtained from the threshold value of τ. Thus, we know the value of the potential at the intersection, $H(R_0) \approx 18$ eV, and we can claculate F_1 for all energies. Therefore, the free parameters in the differential cross section (19) are $x(E)$, $|\dot{\theta}_0|(E)$, and $b(l_0)$.

At E_{lab} = 400 eV, for different values of x from 0.9 to 0.1 and $0 < \lambda \leq 9$, we obtained $I(\theta - \theta_0)$ curves in the region $1° \leq \theta - \theta_0 \leq 10°$[3]. Figure 2 shows the dependence of τ of the first period of the oscillations for different values of x. It is seen from this figure that the experimental period $T \approx 2°$ (see footnote 2) is reached when $x < 0.4$. Inasmuch as the minimum amplitude of the experimentally observed oscillations does not vanish, the best value will apparently be x = 0.14, when the experimental period is reached at $\lambda \approx 7.8$. The ratio of the first maximum to the second is here ≈ 1.05, and the ratio of the first maximum to the first minimum is ≈ 2.9, whereas experiment yields values ≈ 1.5 and ≈ 1.8 respectively (see footnote[2]). Since these ratios are determined mainly by the pre-

FIG. 2. First period of the oscillations of the inelastic cross section (formula (19)) vs. the parameter λ at different x = F_2/F_1 (scattering of He^+ by Ne).



[2]F. T. Smith, Private Communication.

[3]Since we are working in the region $\bar{\tau} < 4{,}000$ eV-deg, it is not necessary to calculate $I(\theta - \theta_0)$ at large values of the angles $(\theta - \theta_0)$.

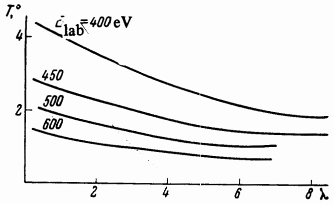


FIG. 3. First period of the oscillations of the inelastic cross section (formula (19)) vs. the parameter λ at different energies (scattering of He^+ by Ne).

exponential function, which we have taken at the point l_0 when integrating with respect to l, such a discrepancy is not surprising. For $x > 0.14$, the experimental period is reached at smaller values of λ, and the ratio of the maximum value of the amplitude to the minimum one will be larger.

Since $F_1 \approx 370$ eV/a when $E_{lab} = 400$ eV, we obtain for $x = 0.14$ that $F_2 \approx 52$ eV/a. But then it follows from the definition of F_2 (see formula (5)) that $dH_{22}/dR_0 \approx 280$ eV/a. Thus, a transition takes place into an excited state, whose electronic term at the point of intersection corresponds to attraction. Knowing now dH_{22}/dR_0, we can find the value of F_2 for different energies. For E_{lab} = 400, 450, 500, and 600 eV we calculated the functions $I(\theta - \theta_0)$ in the region $1° \leq \theta - \theta_0 \leq 10°$ at $0 < \lambda \leq 9$.[4)] The dependence of the first period of the oscillations on λ for different values of E_{lab} is shown in Fig. 3. It is seen from the figure that with increasing energy the value of λ at which the experimental period is obtained decreases. Therefore, the ratio of the maximum to the minimum should increase with increasing energy.

Thus, the qualitative change of the amplitudes and) of the period of the oscillations as a function of the energy is obtained in this model sufficiently well. As to quantitative results, there are many circumstances which can cause errors and an associated discrepancy between the theoretical and experimental curves for the differential cross section. First, the uncertainty in the value of the parameters of the elastic potential and in the position of the point of intersection of the terms R_0. (An analysis of the differential cross section with the aid of formula (19) at an arbitrary choice of the potential would lead to a problem with a large number of free parameters). Second, the approximations made in the calculation of the function $I(\theta - \theta_0)$ (the accuracy of the saddle-point method is not better than 10% in the angle region under consideration, and the error in the calculation of the period is apparently smaller than in the calculation of the absolute values of the function $I(\theta - \theta_0)$). At large values of the reduced angle $\tau = \theta E$, the differential cross section of the inelastic scattering increases. This can be the result of the fact that a new inelastic channel is opened, the influence of which on the scattering in the channel under consideration is disregarded. Nonetheless, when $\tau \leq 4000$ eV-deg, formula (19) apparently describes the experiment satisfactorily, and its analysis can serve as a source of information concerning the inelastic-potential parameters.

In conclusion, the author is sincerely grateful to E. E. Nikitin for suggesting the topic and continuous interest in the work, and M. Ya. Ovchinnikova for numerous useful discussions.

4) The integral in the function $I(\theta - \theta_0)$ is calculated by the saddle-point method in that region of λ, where the equation for the determination of the saddle point has two real roots. Knowing u_{eff}, we can calculate the parameter of the expansion with respect to l. For F_{lab} = 400, 450, and 500 eV we have $(l - l_0)/l_0 \approx 10^{-2}$. At E_{lab} = 600 eV, this parameter becomes equal to 10^{-1}, but such an increase of $(l - l_0)/l_0$ is apparently connected with the fact that the accuracy with which u_{eff} is determined at $\lambda < 1$ is worse than in the preceding cases, since the saddle points come closer together with decreasing λ. Nonetheless, even at 600 eV energy, the expansion in l in formula (16) can be regarded as justified.

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