NONADIABATIC TRANSITIONS IN SLOW COLLISIONS BETWEEN HEAVY PARTICLES

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Submitted to JETP editor June 1, 1963

J. Exptl. Theoret. Phys. (U.S.S.R.) **46**, 863-871 (March, 1964)

The parametric-method equations are solved in the adiabatic approximation by expanding the solution in powers of the colliding-particle velocities. Asymptotic integration of the differential equation yields the transition probabilities in the presence of a frequency minimum and a formula which is an extension of familiar results of the pseudo-intersection theory but does not possess its shortcomings. The method can be used to investigate the effect of other features of the interaction and frequency ω on the cross section for various inelastic transitions in the adiabatic region.

A theoretical investigation of slow collisions between heavy particles in the presence of a point of pseudo-intersection of levels was carried out by Zener [1] and Stueckelberg [2], who obtained an identical result 1). The shortcomings of the Landau-Zener theory were pointed out by Bates [3]. The connection between the two-level approximation with the problem of superbarrier reflection, established by Dykhne and Chaplik, [4,5] has made it possible to obtain a formula analogous to the Stueckelberg formula, valid for the case when the transition region is much smaller than or of the order of the distance between the zeroes of the function ω; this corresponds to very low colliding-particle velocities.

The Landau-Zener theory was made more precise in many papers (for example [6]) by different model approximations of the interaction matrix element. However, owing to the strong dependence of the theory on the analytic properties of the interaction and on the frequency, the question of the applicability of these formulas to specific problems remains open. This circumstance was already pointed out before [7].

In the present paper asymptotic integration is carried out of the equations of the parametric method, and a general formula is obtained for the probability of a double transition in the presence of a minimum of the frequency ω. From this integration it is possible to obtain the results of Stueckelberg, Dykhne, and Zener as limiting cases, and also the general dependence of the transition probability on the parameters that characterize the point of minimum of ω and the velocity in a wider velocity interval. It must be pointed out that the character of the obtained adiabatic solutions makes it possible to trace more fully the connection between the parametric method and the wave method, connection between the coming together of the two terms and the problem of over-the-barrier reflection, and also the characteristic features of symmetrical resonance.

For simplicity we solve the Schrödinger equation by the parametric method, assuming the trajectories of the nuclei to be straight lines. For most problems such an approximation is permissible. Then $z = vT$, where v is the velocity of the colliding particles, T the time, and z the projection of the ray's vector R on the z axis. The system of equations for the transition coefficients, based on the expansion of the total wave function in a series in the atomic wave functions φ_j, can be written in the form ($\hbar = 1$):

$$i\frac{da_k}{dz} = \sum_{j\neq k} \frac{V_{kj}}{v} \exp\left(\frac{i}{v}\int^{z} \alpha_{kj}\, dz'\right) a_k . \qquad (1)$$

For simplicity we assume that the functions φ_j are orthogonal. If they are not orthogonal (charge exchange), the differences are not of fundamental significance [8]. In some problems we can confine ourselves to the two-level approximation. Then the system of equations for the transition coefficients takes the form

$$i\frac{da_{1,2}}{dz} = \frac{V}{v} \exp\left(\pm\frac{i}{v}\int^{z} \alpha dz'\right) a_{2,1}, \qquad (2)$$

$$|a_1(-\infty)| = 1, \quad a_2(-\infty) = 0, \qquad (2a)$$

1) A more general formula for the transition probability in the form of a function that depends on integrals along a complex contour, which go around the zeroes of ω, is the direct consequence of the method of phase integrals (see the Appendix).

where $V = H_{12}$ is the atomic matrix element of the interaction, $\alpha = H_{11} - H_{22}$ is the difference in the terms of first-order perturbation theory. The system (2) was recently considered in [9], but was not integrated there, and only the phases of the asymptotic solutions were used. Consequently the formula obtained in [9] yields the same result as the adiabatic perturbation theory in the case of practical interest when ω has a minimum.

By reducing the system (2) to a single second-order equation for the transition coefficients $a_2(z)$ and by introducing a new function $y(z)$ defined by

$$a_2(z) = V^{1/2} \exp\left(-\frac{i}{2v}\int^z \alpha dz'\right) y(z), \tag{3}$$

we obtain

$$\frac{d^2y}{dz^2} + \frac{\Omega^2}{v^2} y = 0; \tag{4}$$

$$\Omega^2 \equiv \frac{\omega^2}{4} - \frac{iVv}{2}\frac{d}{dz}\frac{\alpha}{V} - v^2\{V,z\}, \quad \omega^2 = \alpha^2 + 4V_2,$$

$$\{V, z\} \equiv \frac{3}{4}\frac{V'^2}{V^2} - \frac{1}{2}\frac{V''}{V}. \tag{5}$$

Here $\{V, z\}$ is the Schwartz derivative of V with respect to z. Equation (4) is analogous to the equation for a wave propagating in a medium with complex refractive index.

We seek a solution of (4) for low velocities in the form [2)]

$$y(z) = \exp\left\{\frac{1}{v}(S_0 + vS_1 + v^2S_2 + \ldots)\right\}. \tag{6}$$

Substituting the expansion (6) in (4) and equating coefficients of equal powers of v, we obtain, accurate to terms proportional to v in the argument of the exponent, an approximate solution for (4) in the form

$$y(z) = \frac{A_+}{\sqrt{\omega}}e^{iQ} + \frac{A_-}{\sqrt{\omega}}e^{-iQ}; \quad Q = \frac{1}{2v}\int_{z_1}^{z}\omega dz' - \frac{i}{2}\int_{t_1}^{t}\frac{dt'}{\sqrt{1-t'^2}}$$

$$t = \frac{\alpha}{2V}, \tag{7}$$

where A_+ and A_- are constants. In this form the solution is very similar to the quasiclassical wave function of Stueckelberg [[2], formula (26)], the only difference being that in [2] we deal with two waves that "propagate" along two reaction channels.

From the boundary conditions (2a) we have

$$A_+ = 0. \tag{8}$$

The choice of the coefficient A_- depends on the start of the integration and on the argument of the exponent, and is determined by a condition that follows from the system (2), viz.,

$$\left|\frac{da_2}{dz}\right| \to \left|\frac{V}{v}\right| \quad \text{as} \quad z \to -\infty. \tag{9}$$

Differentiating (3) with allowance for (7) and (8) and then substituting the result in (9), we obtain for the coefficient A_- the following expression (the start of integration in the exponent is the point $z = z_1$)

$$|A_-| = (1 + k^2/4)^{-1/2}(t_1 + \sqrt{1 + t_1^2})^{-1/2}, \tag{10}$$

$$k = 2pv/\Delta E, \quad p = V'(-\infty)/V(-\infty). \tag{11}$$

Under condition (8), the function (7) tends to zero as $z \to +\infty$ if we move along the real z axis, and we obtain no transition whatever. Inclusion of the next terms of the expansion (6) does not change the situation, that is, a complete analogy is obtained here with the problem of over-the-barrier reflection in the quasiclassical approximation. For symmetrical resonance $\alpha = 0$; if we choose $A_+ = A_-^*$, we obtain for the transition probability the well-known formula

$$P = \sin^2\left(\frac{1}{v}\int_{-\infty}^{+\infty} Vdz'\right). \tag{12}$$

We thus obtain for the symmetrical process a nonzero transition probability due to the adiabatic transitions. The expansion (7) can obviously not be employed near the zeroes and singularities of the function ω. We confine ourselves henceforth to an investigation of the very important case when ω has a minimum at the points $z = z_1$ and $z = z_2$, and consequently it has a pair of complex-conjugate roots near each of these points in the complex z plane. We neglect the influence of the remaining zeroes and singularities of ω and V, which are situated far from the real axis.

At low colliding-particle velocities, determined by the condition for discarding the terms of order v in the exponent, we can continue the solution (7) with boundary conditions (8) and (10) from the region $z \ll z_1$ into the region $z \gg z_2$ by the phase-integral method. As shown in the appendix, we have in this case the following transition formula:

$$A_-\omega^{-1/2}e^{-iQ(z)} \to -2iA_-\omega^{-1/2}e^{\nu}(1 - e^{2\nu})^{1/2}\sin\mu$$

$$\exp\{i[Q(z) - Q(z_2)]\}, \tag{13}$$

$$\nu = \frac{i}{4v}\oint^{(+)}\omega dz', \quad \mu = \frac{1}{2v}\int_{z_1}^{z_2}\omega dz'. \tag{13'}$$

The contour in the integral for ν is drawn clockwise around the roots of ω. Recognizing that

$$\int^t \frac{dt}{\sqrt{1+t^2}} = \ln(t + \sqrt{1+t^2}) + C$$

2) The small parameter is essentially the quantity $\sqrt{h}$.

and that $t \to \infty$ as $z \to \infty$, we obtain for the transition probability

$$P = 4K(k)\, e^{2\nu}(1 - e^{2\nu}) \sin^2 \mu; \qquad (14)$$

$$K(k) = (t_1 + \sqrt{1 + t_1^2})^{-2} (1 + k^2/4)^{-1}, \quad k = 2pv/\Delta E. \qquad (15)$$

Equation (14) coincides with the Stueckelberg formula apart from the term K, which has not been taken into account. For the reaction with charge exchange, p is a quantity proportional to the smallest ionization potential. Thus, the term K appears naturally as a consequence of allowance for the time variation of V and the fact that the terms do not move apart at infinity.

In order to obtain a formula which is valid in a broader velocity interval, and also to estimate the region of applicability of (14), we present a more accurate method for integrating (4) with the aid of the associated equation. This method is not connected directly with the expansion of (7), and if ω and V are smooth it can give a result which is valid over a wider velocity interval.

If the following inequality is satisfied on any contour around two zeroes of the function ω,

$$\frac{|\omega^2|}{4} > v \left| \frac{iV}{2} \frac{d}{dz} \frac{\alpha}{V} + v\{V, z\} \right|, \qquad (16)$$

then on the basis of a known theorem concerning the roots of analytic functions we can state that Ω also has two roots in this region $\Omega \to \omega/2$ as $v \to 0$ and the roots of Ω coincide with the roots of ω. Inequality (16) imposes a limitation on the range of variation of the velocities.

We denote the roots of Ω near the point z_1 by b_1 and b_2 and near the point z_2 by b_3 and b_4. We now choose in the vicinity of the point $z = z_1$ an associated equation with a coefficient that has two zeroes. The theory of the associated equation can be found in papers by many authors [10-12]. By way of such an equation we choose the equation for the parabolic-cylinder functions:

$$d^2U/dx^2 + (x^2/4 + \nu)\, U = 0. \qquad (17)$$

In order to reconcile the roots of Ω and $x^2/4 + \nu$ and to obtain the correct asymptotic behavior at large values of these quantities, the connection between the variables z and x must be established in the form

$$\frac{1}{v} \int_{b_1}^{z} \Omega dz' = \frac{1}{2} \int_{-2i\sqrt{\nu}}^{x} \sqrt{x^2 + 4\nu}\, dx' \qquad (18)$$

and the parameter ν must be chosen from the condition

$$\frac{1}{v} \int_{b_1}^{b_2} \Omega dz' = \frac{1}{2} \int_{-2i\sqrt{\nu}}^{+2i\sqrt{\nu}} \sqrt{x^2 + 4\nu}\, dx' = i\pi\nu. \qquad (19)$$

Putting

$$\Phi(z) = \frac{1}{v} \int_{z_1}^{z} \Omega dz', \qquad (20)$$

$$\Phi_1 = \frac{1}{2v} \int_{z_1}^{b_1} \Omega dz' + \frac{1}{2v} \int_{z_1}^{b_2} \Omega dz', \qquad (21)$$

we can write the substitution (18) in the form

$$\Phi - \Phi_1 = \frac{1}{2} \int_0^x \sqrt{x^2 + 4\nu}\, dx' = \frac{1}{4} x \sqrt{x^2 + 4\nu} + \nu \ln (x + \sqrt{x^2 + 4\nu}) - \frac{\nu}{2} \ln 4\nu. \qquad (22)$$

When z varies along the real axis from $-\infty$ to ∞, x changes along a complex contour whose ends go to infinity.

We assume, for concreteness, that $\alpha > 0$ when $z < z_1$ and $z > z_2$, and choose a branch of the function Ω such that $\mathrm{Re}\, \Omega > 0$ when z is real. In the approximation under consideration the solutions of (4) and (17) are connected by the relation

$$y(z) = \sqrt{dz/dx}\, U(x). \qquad (23)$$

The solutions of (17) will be the parabolic-cylinder functions

$$U_{1,2}(x) = D_{\pm i\nu - 1/2}(e^{\mp i\pi/4} x). \qquad (24)$$

The asymptotic expressions for the functions $U_1(x)$ and $U_2(x)$ at large values of x, when $\mathrm{Re}\,[\exp(\pm \pi i/4)x] < 0$, are of the form

$$U_{1,2} \to \exp\left(\pm \frac{ix^2}{4} + \frac{\pi\nu}{4} \pm \frac{\pi i}{8}\right) x^{\pm i\nu - 1/2}, \qquad (25)$$

and when $\mathrm{Re}\,[\exp(\pm \pi i/4)x] < 0$, they are obtained with the aid of the well-known turning formulas (see, for example, [13]).

Using (22), we can write

$$U_1(x) \to \exp\left(\frac{\pi\nu}{4} + \frac{\pi i}{8} + \frac{i}{2}\nu \ln \nu - \frac{i\nu}{2}\right) (x^2 + 4\nu)^{-1/4} \times \exp\left(\frac{i}{2} \int_0^x \sqrt{x'^2 + 4\nu}\, dx'\right), \qquad (26)$$

$$\mathrm{Re}\,(e^{-i\pi/4} x) > 0. \qquad (27)$$

The corresponding expression for $U_2(x)$ is obtained by replacing i with $-i$. Equation (4) is solved in the same manner near the point $z = z_2$, the only difference being that Φ_1 is replaced here by

$$\Phi_2 = \frac{1}{2v} \int_{z_2}^{b_3} \Omega dz' + \frac{1}{2v} \int_{z_2}^{b_4} \Omega dz' \qquad (28)$$

and $\nu_2 = \nu_1^* = \nu^*$, where ν_1 and ν_2 are the values of the parameter ν at $z = z_1$ and z_2.

Near the point $z = z_1$ we represent the solution in the form

$$y(z) = N_- \sqrt{\frac{2}{v}\frac{dz}{dx}}\, U_1(-x), \tag{29}$$

and near $z = z_2$ in the form

$$y(z) = \sqrt{\frac{2}{v}\frac{dz}{dx}}\, [M_1 U_1(-x) + M_2 U_2(-x)]. \tag{30}$$

After simple but rather long and cumbersome calculations, we obtain the following formula for the transition from the region $z \ll z_1$ to the region $z_1 < z < z_2$ and then to the region $z \gg z_2$:

$$N_- \Omega^{-1/2} e^{-i\Phi(z)}$$
$$\to N_- \Omega^{-1/2}\left[e^{i(\Phi - 2\Phi_1 - \pi/2 + i\pi\nu)} + \frac{1}{\Gamma(1/2 - i\nu)} e^{-i(\Phi + \nu\ln\nu - \nu - i\pi\nu/2)}\right]$$
$$\to \frac{\sqrt{2\pi} N_-}{|\Gamma(1/2 + i\nu)|}\Omega^{-1/2}\left[e^{i\Phi(z_2) + i\varphi} - e^{-i\Phi(z_2) - i\varphi}\right] e^{i[\Phi(z) - \Phi(z_2)]}, \tag{31}$$

where the phase φ is expressed in rather complicated fashion in terms of the phases of ν and γ-functions. We shall not write out the phase since the term in the square brackets averages out in the final results.

The transition formula is obviously compatible with the boundary condition (2a), and the coefficient N_- is again determined from the condition (9). We obtain ultimately for the transition probability

$$P = \frac{2\pi K_1(k)}{|\Gamma(1/2 + i\nu)|}\left|e^{(-3\pi\nu/2 + i\nu\ln\nu - i\nu)}\right|^2 \left|e^{i[\Phi(z_2) + \varphi]} - e^{-i[\Phi(z_2) + \varphi]}\right|^2; \tag{32}$$

$$K_1(k) = 8\,[2 + k^2/2 + 2\sqrt{1 + k^2} + 2\sqrt{2}(\sqrt{1 + k^2} + 1)^{1/2} + \sqrt{2}k(\sqrt{1 + k^2} - 1)^{1/2}]^{-1}(t_1 + \sqrt{1 + t_1^2})^2,$$

$$\nu = \frac{1}{2\pi i v}\oint^{(-)} \Omega dz', \quad k = \frac{2pv}{\Delta E}, \tag{33}$$

and we take the integral over a closed counterclockwise contour around the roots.

If the simple inequality (16) is replaced by a strong inequality, we can write approximately

$$\Omega \approx \frac{\omega}{2} - \frac{iv}{2}\frac{dt/dz}{\sqrt{1 + t^2}}. \tag{34}$$

We then obtain

$$\nu = \tau_1 - i\tau_2/2, \tag{35}$$

where

$$\tau_1 = -\frac{i}{4\pi v}\oint^{(+)} \omega dz', \quad \tau_2 = 1, \quad \omega\,|_{z<z_1} > 0. \tag{36}$$

We obtain for the transition probability in this case

$$P = 4K_1 e^{-2\pi\tau_1}(1 - e^{-2\pi\tau_1})\exp\left(2\tau_1 \tan^{-1}\frac{1}{2\tau_1} - 1\right) \times \sqrt{1 + \frac{1}{4\tau_1^2}}\,\sin^2\left(\frac{1}{2v}\int_{z_1}^{z_2}\omega dz' + \varphi\right). \tag{37}$$

If we assume that $\tau_1 \gg 1$, then we have

$$P = 4K_1 e^{-2\pi\tau_1}(1 - e^{-2\pi\tau_1})\sin^2\left(\frac{1}{2v}\int_{z_1}^{z_2}\omega dz'\right). \tag{38}$$

This coincides with formula (14), which is equivalent to the Stueckelberg formula apart from a factor K_1. If we replace the square of the sine by $1/2$, we obtain the formula of Dykhne and Chaplik[5] (accurate to K_1).

If we use the expansion near z_1

$$\omega^2 = \omega_1^2 + \frac{\omega_1^{2'}}{2}(z - z_1)^2, \tag{39}$$

then we obtain in this approximation

$$P = 2K_1 e^{-v_0/v}(1 - e^{-v_0/v}), \tag{40}$$

$$v_0 = \pi\omega_1^2/\sqrt{2}\sqrt{\omega_1^{2''}}, \tag{41}$$

where $K_1 \approx k$ if k is small.

If the terms cross, then $\alpha_1 = 0$, $t_1 = 0$, and $v_0 = 2\pi V_1^2/\alpha_1'$, that is, (40) coincides with the Zener formula (accurate to K).

If the terms do not cross, we have approximately

$$v_0 = \pi(\alpha_1^2 + 4V_1^2)/4V'\sqrt{1 + \alpha'^2/4V'^2}. \tag{42}$$

It must be noted, however, that (38) and (40) are obtained only if $|\nu| \gg 1$, when the second term in the parentheses is essentially small. Thus, they are not applicable in the region of the maximum probability, when $|\nu|$ is of the order of magnitude of unity. In this case it is necessary to use the more general formula (32) or the simplified formula (37), which are valid in a wider range of velocities, determined by the inequality (16), and which give the correct dependence of the transition probability on the system parameters.

It can be verified that the transition probability determined by (32) [or by the simplified (37)] does not exceed unity; at very small v, (37) coincides with the Zener formula, and at large v it gives a decrease like v^{-2}, something missing from the Zener theory.

For the reactions $A^{n+} + B \to A^{(n-1)+} + B^+$ (see the papers of Bates and his co-workers[14-16]) we have in the approximation given by (37) the following expression for the charge-exchange cross section:

$$\sigma = 4\pi R_x^2 K_1(k) f I_1(\eta), \tag{43}$$

where R_x is the distance to the point of pseudo-intersection, f the statistical weight of the corresponding term, and

$$I_1(\eta) = \int_1^\infty e^{-\eta x}(1 - e^{-\eta x})\sqrt{1 + \frac{\pi^2}{(\eta x)^2}} \times \exp\left\{\frac{\eta(x)}{\pi}\tan^{-1}\frac{\pi}{\eta x} - 1\right\} x^{-3} dx,$$

$$\eta = 247\,(n-1)\,\mu^{1/2}\,[\Delta U_{R_x}/\Delta E]^2 E^{-1/2}, \qquad (44)$$

where μ — reduced mass in O^{16} units, ΔU_{R_x} — separation of the terms, ΔE — resonance defect, and E_p — energy of relative motion in eV. We must take here I_B for p.

We did not use any model representation in our deduction, and took account only of the presence of zeroes of ω which lie close to the real z axis; these zeroes determine the region of non-adiabatic transitions. Consequently our derivation is free of the shortcomings of the Zener theory, connected with the nonseparation of the terms at infinity, failure to take into account the time variation of the matrix element, the breakdown of the perturbation into two parts, etc. The analogy with the over-the-barrier reflection and the connection between the adiabatic approximation and the quasiclassical theory is most completely manifest here.

In conclusion we note that the phase-integral or associated-equation methods can be used to investigate the influence of other analytic singularities of ω and V, which determine the transition probabilities, and consequently also the cross sections for the different inelastic processes in the adiabatic region.

We take the opportunity to thank Yu. N. Demkov for a discussion of the work.

APPENDIX

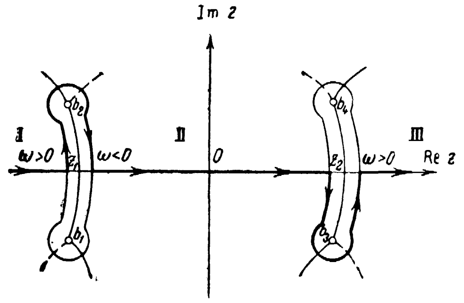
The method of phase integrals is based on the fact that if a function y(z) is analytic, then this property is not possessed by its asymptotic expansions, and a jumplike change in the coefficients (the Stokes phenomenon) is necessary in order to represent the function with the aid of its asymptotic expansions on the entire plane. This jumplike variation of the coefficients occurs on Stokes lines which are determined for our case by the condition

$$\arg\left(\pm i\int_{b_j}^{z}\omega dz'\right) = \pi + 2n\pi, \quad j = 1, 2, 3, 4. \qquad (A.1)$$

These lines for A_+, shown solid in the figure, leave the points b_1 and b_2 at angles $2\pi/3$ and $5\pi/3$, while the lines for A_1 leave at angles $\pi/3$ and $4\pi/3$ (they are shown dashed).

For the point z_2 the situation is reversed, for whereas we have $\omega > 0$ for $z < z_1$ when z is real, we obtain $\omega < 0$ for $z_1 < z < z_2$. The standard procedure for determining the Stokes coefficients yields in this case[2]

$$\alpha = \beta = \gamma = \delta = e^{i\pi/2}(1 - e^{2\nu})^{1/2}, \quad \nu = \frac{i}{4v}\oint^{(+)}\omega dz'. \qquad (A.2)$$



The continuation of the solution (7), with allowance for (8), along the contour shown in the figure, from the region I into region II and then into region III, is realized in the following fashion:

$$A_-\omega^{-1/2}e^{-iQ(z)} \to \alpha A_- M_1 M_2 \omega^{-1/2} e^{iQ(z)} + (1+\alpha\beta)\, A_- M_2^2 \omega^{-1/2} e^{iQ(z)}$$

$$\to [\alpha A_- M_1 M_2 N_1^2 e^{-i\mu}$$

$$+ \alpha(1+\alpha\beta)\, A_- M_2^2 N_1 N_2 e^{i\mu}]\, \omega^{-1/2} e^{iQ(z)-iQ(z_2)}$$

$$+ B_-\omega^{-1/2} e^{-iQ(z)+iQ(z_2)}; \qquad (A.3)$$

$$\mu = \frac{1}{2v}\int_{z_1}^{z_2}\omega dz', \quad M_1 = \exp\left(\frac{i}{2v}\int_{z_1}^{b_2}\omega dz' + \int_{t_1}^{t(b_2)}\frac{dt}{\sqrt{1+t^2}}\right),$$

$$N_1 = \exp\left(\frac{i}{2v}\int_{z_2}^{b_3}\omega dz' + \frac{1}{2}\int_{t_2}^{t(b_3)}\frac{dt}{\sqrt{1+t}}\right),$$

$$M_2 = M_1^{-1}, \quad N_2 = N_1^{-1}, \qquad (A.4)$$

and the value of the coefficient B_- will not be written out, since this term vanishes when $z \to +\infty$. Using the relations

$$M_1 M_2 = N_1 N_1 = 1, \quad N_1^2 = M_2^{2*} = e^{(\nu - i\pi)/2}, \qquad (A.5)$$

we obtain in this case formula (13) of the main text for the transition from region I into region III.

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Translated by J. G. Adashko
127