CHARGE EXCHANGE IN COLLISIONS OF MULTIPLY CHARGED IONS

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Submitted to JETP editor December 22, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 48, 1499-1507 (May, 1965)

The probability of charge exchange of multiply charged ions ($A^{+2} + B \rightarrow A^{+} + B^{+}$) is calculated, within the framework of the theory of nonstationary perturbations and in the two-level and classical nuclear-motion approximations for the case when there is Coulomb interaction in one of the channels ($A^{+} + B^{+}$). The nondiagonal interaction matrix element in the atomic-function representation is approximated by an exponential function of the interatomic distance. The system of time-dependent equations for the complex transition amplitudes, written in the representation of molecular electronic functions, is integrated numerically for a wide range of variation of the parameters of the problem (δ and γ). The limits of applicability of the Landau-Zener formula are delineated.

1. FORMULATION OF THE PROBLEM

THE calculation of the cross section for charge-exchange in atomic collisions involves two problems. The first is to find approximate adiabatic electronic wave functions of the system of two atoms. In those cases when the charge exchange (that is, the transition between two adiabatic electronic states) occurs essentially at large internuclear distances R and is considered as a single-electron process, the adiabatic functions can be approximated with sufficient degree of accuracy by a linear combination of atomic orbitals[1-4], with a pre-exponential factor suitably corrected for the distortion of the atomic orbitals in the field of the other center.[5] We shall henceforth consider precisely this case, and the lower bounds of the quantity R will be indicated later on.

The second problem is to solve the coupled Schrödinger equations for the wave functions of the relative motion of the nuclei, or a system of time-dependent equations in which account is taken of the interaction induced between the adiabatic electronic states by the motion of the nuclei, and of other terms left out of the adiabatic electronic Hamiltonian. We retain in these equations only two terms (between which a transition takes place), and assume that the motion of the nuclei is classical and that the trajectory of motion $R = R(t)$ is determined by an adiabatic potential. This imposes additional limitations on the splitting ΔE of the adiabatic terms and on the energy ϵ of relative motion of the partners in the collision.

These limitations can be easily obtained from an analysis of the potential curves of the terms between which the transition takes place. Let us consider for concreteness a reaction of the type (11/20) [1]:

$$A^{+} + B^{+} \rightleftarrows A^{+2} + B + \Delta\varepsilon. \qquad (1)$$

We assume (as will be confirmed subsequently) that the main contribution to the probability of transition between the two terms in question comes from a certain region with center R_0 and effective width ΔR. Then, if the condition $(\Delta R/R_0) \ll 1$ is satisfied, the system develops in the internal region ($R \lesssim R_0$) adiabatically, so that there are no transitions near the turning points. In the region of the transition ($R \sim R_0$) we can introduce the concept of a trajectory provided $\Delta E(R_0)$ is much less than ϵ_1 or ϵ_2. In addition, the energies ϵ_1 and ϵ_2 are bounded from above; this limitation follows from the requirement that the two states in question be weakly coupled with all other states, and can be approximately formulated in the form

$$\left| \sum_{n \neq i} (A_{in}/\Delta E_{in}) \right| \ll 1 \quad (i = 1, 2),$$

where ΔE_{in} is the difference between the energies of the adiabatic terms i and n while A_{in} is the matrix element of the nonadiabatic interaction between these states (Fig. 1).

A characteristic feature of the reaction (11/20) is that the difference of the terms ap-

[1] The notation is that of Hastead (see[4], page 696). The numbers indicate the charges of the two initial and the two final ions.

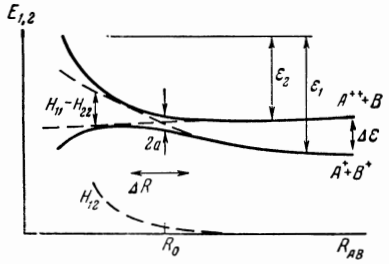

FIG. 1.

proaches the asymptotic splitting $\Delta\epsilon = \Delta E(\infty)$ like $1/R$ (we are using atomic units throughout). This leads to the possibility of crossing of the zeroth-approximation terms (that is, the terms calculated with atomic functions). At such large distances $R = R_0$, so that we can use for the non-diagonal matrix element of the interaction the asymptotic expression

$$V(R) \approx CR^n e^{-\alpha R}, \quad \alpha = \sqrt{2I},$$
$$I = \min(I_B,\ I_A^+,\ I_B^+). \tag{2}$$

C and n can be obtained by the method given in Smirnov's paper[5] (see also [1-3]).

Approximation (2), the form of which is in general important only in the region of the transition, is valid if $(\Delta R/R_0) \ll 1$. In a more rigorous approach it would be necessary to use as the pre-exponential function a polynomial in R of degree n, but when $\alpha R \gg 1$ the rate of growth of this polynomial is determined only by the principal term $(\alpha R)^n$. Since as a rule $\Delta R \approx 1/\alpha$, the condition $(\Delta R/R_0) \ll 1$ (which includes the requirement $\alpha R_0 \gg 1$) is satisfied [2)] if $\alpha R \gg n$. But this means that the pre-exponential factor in (2) can be regarded in the transition region as constant, that is, we can put in final analysis

$$V(R) \approx be^{-\alpha R}.$$

Thus, if the following conditions are satisfied

$$\Delta R/R_0 \ll 1, \quad \Delta E(R_0) \ll \varepsilon_1,\ \varepsilon_2,$$
$$\left| \sum_{n \neq i} (A_{in}/\Delta E_{in}) \right| \ll 1 \tag{3}$$

then reaction (1) is described by a system of two equations for the amplitudes a_1 and a_2 of the expansion of the total wave functions in the atomic orbitals φ_1 and φ_2 or for the amplitudes b_1 and b_2 of the expansion in the molecular functions ψ_1 and ψ_2. Within the framework of the approximation of the two states, this system of equations can be regarded as Hermitian, since the nonhermiticity due to the non-orthogonality of the atomic functions is brought about only by the influence of the higher electronic states, which are neglected here. Without stopping to discuss this question in greater detail, we note only that the nonhermiticity can be eliminated by using, for example, a single-center basis, at the expense of formulating the initial conditions somewhat inaccurately[6].

As a model interaction illustrative of the nature of the problem, we use the following matrix elements, calculated in the basis of atomic functions (which are orthogonal by definition)

$$H_{11} - H_{22} = \Delta\varepsilon - 1/R \approx (R - R_0)/R_0^2 + \text{const},$$
$$H_{12} = a \exp(-\alpha(R - R_0)). \tag{4}$$

In the region of the transition we approximate R by a linear function of the time, $R = R_0 + v_R d$, choosing as the origin the point $R = R_0$, where $H_{11} = H_{22}$. Thus, the parameters of the model are as follows: the difference ΔF in the strengths of the linear terms, the matrix element a of the interaction between the states, the resonance defect $\Delta\epsilon$ of reaction (1) (connected with ΔF by the relation $\Delta F = 1/R_0^2 = \Delta\epsilon/R_0$), and the radius of action $1/\alpha$ of the exchange forces (all the parameters are taken at the point $R = R_0$). It is convenient to define the following dimensionless parameters:

$$\gamma \equiv \hbar v_R \Delta F/2a^2, \quad \delta \equiv 2a\alpha/\Delta F, \quad \tau \equiv at/\hbar. \tag{5}$$

Turning to [1-3], we can establish that the regions of variation of the parameters are as follows

$$\delta = 10^{-4} - 10, \quad \alpha R_0 = 5 - 20. \tag{6}$$

As to γ, this parameter changes in an interval that contains the point $\gamma = \pi/\ln 2 \approx 4.53$ of the maximum transition probability, calculated in the Landau-Zener approximation (H_{12} = const.).

Finally, the system of equations for the transition amplitudes takes the form

$$ida_{1,2}/d\tau = \exp(-\delta\gamma\tau \pm i\gamma\tau^2)\, a_{2,1}. \tag{7}$$

This system describes the shift of the atomic

[2)] One might think that the condition $\Delta R/R_0 \ll 1$ contains the velocity of the colliding atoms. This is actually the case if ΔR is estimated on the basis of the Landau-Zener model: $\Delta R \approx \Delta R_1 = (\hbar v/\Delta F)^{1/2}$. For such an estimate it is assumed, however, that the width ΔR_1 of the transition is much smaller than the characteristic interaction radius $1/\alpha$ (approximation of the matrix element of the interaction by a constant). Therefore it is necessary to substitute in the criterion $\Delta R/R_0 \ll 1$ the value $\Delta R \sim 1/\alpha$, and not $\Delta R \sim R_1$. If it turns out formally that $\Delta R_1 \gg 1/\alpha$, this means only that the width of the transition region cannot be estimated within the framework of the Landau-Zener approximation. From (5) we see that $\Delta R_1 \alpha \approx \delta\gamma^{1/2}$, and when $\delta\gamma^{1/2} \gtrsim 1$, as follows from the results of the paper, the transition probability differs greatly from the value calculated by the Landau-Zener formula (see Fig. 3 below).

states after a single passage through the transition region (approach of the atoms to the turning point). No account is taken here, however, of the time dependence of the atomic functions due to the parametric relation $I = R(t)$ (dynamic coupling). Neglect of dynamic coupling is valid under the condition that the velocity of the nuclei be small compared with the velocity of the atomic electron; this ensures that the factor $\exp(-im_e v_R r/\hbar)$, which is introduced into the atomic weight function to take into account the motion of the nucleus[7], is close to unity. In dimensionless parameters, this condition is approximately formulated in the form $\gamma\delta \ll I/a$. It must be added to the limitations (3) on the velocity of the nuclei.

An investigation of the system (7) is of interest, in particular, because the model on which it is based is analogous to the model considered by Nikitin[8,9] and by Demkov[10] in connection with a charge exchange reaction of the type (01/10). The only difference is that for the charge exchange (01/10) it is necessary to put $\Delta E = \Delta\epsilon = \text{const}$, that is, $i\gamma\tau^2$ in (7) must be replaced by $i\gamma\tau$. It will be shown below that the strong approach of the terms in the reaction (11/02) can lead to a nonmonotonic dependence of the probability of the charge exchange W, and also of the cross section σ, on the relative velocity v_∞ of the atom; in this connection, the results obtained will be compared with the results of some theoretical papers[11-14].

2. GENERAL PROPERTIES OF THE TRANSITION PROBABILITY

We assume first that the difference in the actions along the trajectories from the turning point (R_1 or R_2) to the transition region (R_0) is large, that is,

$$\Delta S \equiv \int_{R_1}^{R_0} \frac{P_1}{\hbar} dR - \int_{R_2}^{R_0} \frac{P_2}{\hbar} dR \approx \int_{R_2}^{R_0} \frac{\partial P}{\partial E} \Delta E\, dR \gg 1. \quad (8)$$

In this case the interference of the transition amplitudes, due to the double passage through the transition region, can be neglected, and the transition probability W for the double passage can be determined simply by summing the currents: $W = 2P(1 - P)$. The probability P is determined by integrating the system of equations (7) as the square of the modulus of the nondiagonal element of the scattering matrix that relates the adiabatic states on the two sides of the transition region. The transformation from the atomic orbitals φ_i to molecular orbitals ψ_i is defined in usual fashion:

$$\begin{pmatrix}\psi_1\\ \psi_2\end{pmatrix} = \begin{pmatrix}\cos\mu & \sin\mu\\ -\sin\mu & \cos\mu\end{pmatrix}\begin{pmatrix}\varphi_1\\ \varphi_2\end{pmatrix}, \quad \tan 2\mu = \frac{2H_{12}}{H_{11} - H_{22}}. \quad (9)$$

The amplitudes b_i (see above) satisfy in this case the following system of equations:

$$\frac{idb_{1,2}}{d\tau} = \dot{\mu} \exp\left(\pm i\int_0^\tau \Delta E\, d\tau'\right) b_{2,1}, \qquad |b_1(+\infty)|^2 = 1,$$

$$b_2(+\infty) = 0, \qquad |b_2(-\infty)|^2 \equiv P; \quad (10a)$$

$$\Delta E(\tau) \equiv 2\int_0^\tau [\gamma^2\tau'^2 + \exp(-2\delta\gamma\tau')]^{1/2} d\tau',$$

$$\dot{\mu} \equiv \frac{\gamma}{2}(1 + \delta\gamma\tau)\frac{e^{-\delta\gamma\tau}}{\sqrt{\gamma^2\tau^2 + e^{-2\delta\gamma\tau}}}. \quad (10b)$$

A discussion of the general properties of P is best carried out in connection with the analytic properties of the function ΔE, which is regarded as a function of the complex variable $z \equiv \delta\gamma\tau$, inasmuch as in the case of small velocities ($\Delta E_{min}/\alpha\hbar v \gg 1$) the main contribution to the transition probability P is made by the regions near the branch points of $\Delta E(\tau)$[15]. The zeroes of the radicand in (10b) are determined by the points of intersection of the curves

$$\arg(\pm ize^z) = 0, \quad |\pm ize^z| = \delta, \quad (11)$$

which are shown in Fig. 2. It is easy to determine from the figure the character of motion of the zeroes in the z plane as δ is varied.

When $\delta \ll 1/e$, Eq. (11) describes two curves, one of which is localized near the origin. On these curves lie the two zeroes of ΔE (z_0 and z_0^*) which are closest to the real axis. The second curve does not cross the y axis, and all the branch points on it (z_n and z_n^*) lie farther from the y axis than z_0 or z_0^*. In the limit as $\delta \to 0$, there are actually two independent regions of the transition; the first is situated near $x = 0$ (point of crossing of the zeroth-approximation terms), and the second is near $x = x_0$, where x_0 is the root of

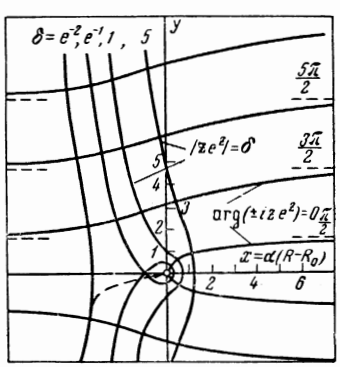

FIG. 2.

the equation $\delta e^{-x} = x$ (the point at which the difference in the zeroth-approximation terms is equal to double the matrix element of the interaction). If the distance between the two indicated transition regions is large, then the transition probability is obtained by summing the currents, that is,

$$W = W_1 + W_2 - 2W_1W_2. \tag{12}$$

In this case W_1 is given by the Landau-Zener formula[16] (two branch points), and we can use for W_2 the result of the calculation of the transition probabilities of two parallel first-approximation terms[9,17] (infinite number of branch points, located at equal distances from one another and lying on a straight line parallel to the imaginary axis):

$$W_1 = 2e^{-\pi/\gamma}(1 - e^{-\pi/\gamma}); \tag{13}$$

$$W_2 = 2e^{-\beta}(1 + e^{-\beta})^{-2},$$

$$\beta \equiv 2\pi x_0(\delta)/\gamma\delta^2, \quad x_0 = \delta e^{-x_0}. \tag{14}$$

The condition for neglecting interference between the transition amplitudes reduces to the condition of smallness of the effective widths of the transition regions compared with the distance between them, that is,

$$x_0 \gg 1, \qquad x_0 \gg \delta\sqrt{\gamma}. \tag{15}$$

It is easy to see that simultaneous satisfaction of these relations results in the inequality

$$\Delta S \approx \int \frac{\Delta E}{\hbar v}\, dR \sim \frac{x_0^2}{\delta^2\gamma} \gg 1. \tag{16}$$

When $\delta \gg 1$, as can be seen from Fig. 2, there remains one transition region, and the transition probability can be represented asymptotically (when $x_0 \gg 1$) by formula (14), inasmuch as the branch points are again located on a line almost parallel to the y axis.

Under conditions (3) it is easy to find the correct phase factor which must be introduced into the expression for the transition probability in the limit of large velocities. In this case the probability W reaches a maximum value, and the phase factor can be obtained within the framework of perturbation theory for degenerate states[18] by assuming that the interaction is turned on in the transition region instantaneously [$(\Delta E_{min}/\alpha\hbar v) \ll 1$] and by joining the adiabatic functions in the internal region ($R \lesssim R_0$) to the atomic functions in the external region ($R \gtrsim R_0$). We thus obtain

$$W' = 2P(1-P)\cdot 2\sin^2\left(\frac{1}{2\hbar}\int \Delta E(R)\,\frac{dR}{v_R}\right). \tag{17}$$

A consequence of (3) is the inequality

$$S = \int\limits_{R<R_0} \frac{\Delta E\, dR}{\hbar v_R} \gg \frac{\Delta\varepsilon}{\alpha\hbar v_R}, \tag{18}$$

and therefore the approximate determination of the phase results in a small error if the Massey parameters are small ($\Delta e/\alpha\hbar v \ll 1$). In other words, we can describe with sufficient accuracy the velocity region corresponding to the monotonic decrease of the reaction cross section (limit of the Born approximation). In the region of large oscillations of the cross section, the method of joining gives an incorrect phase shift, but this phase shift is immaterial, since the observed cross section is a quantity averaged over the oscillations.

3. NUMERICAL INTEGRATION OF THE FUNDAMENTAL SYSTEM OF EQUATIONS

In the intermediate region of the parameter δ ($\delta \sim 1$), the system of four differential equations for the real and the imaginary parts of the transition amplitudes was integrated numerically. The presence of rapidly oscillating coefficients in the system makes the numerical integration most difficult. This is precisely why the initial system (7) cannot be integrated at all in practice. In fact, with increasing $\tau > 0$, both the amplitude $e^{\alpha'\tau}$ and the phase $\beta'\tau^2$ increase rapidly in the coefficients of the type $e^{\alpha'\tau}\sin\beta'\tau^2$ ($\alpha' > 0$). Therefore, in order to maintain the required accuracy of integration (≈ 0.01 in each of the integrated functions) during each step, in the presence of accelerating oscillations of the increasing amplitude, it is necessary to integrate with smaller and smaller intervals. Then the integration time necessary to obtain the asymptotic value of P (as $\tau \to -\infty$) becomes very large. The advantages of the adiabatic representation of (10) lie in the presence of the amplitude $\dot{\mu}$, which exponentially decreases [see (10b)] on both ends of the integration interval[3)] (+A, −A), in front of the rapidly oscillating factor

$$\exp\left(i\int\limits_0^{\tau} \Delta E\, d\tau'\right).$$

By virtue of this, the time connected with the need for calculating the integral

$$\int\limits_{\tau_{n-1}}^{\tau_n} \Delta E\, d\tau'$$

[3)] The limits of integration were determined by trial and error, starting from the requirement that there be no transitions at $\tau = \pm A$, accurate to $\sim 10^{-6}$.

during each n-th step is compensated by the exponentially rapid approach of the solution to the asymptotic value. The numerical integration was by the Runge-Kutta method, with automatic selection of the interval such as to ensure a specified integration accuracy (see above). The region of variation of the parameters δ and γ, and also the results of the calculations, are shown in Fig. 3.

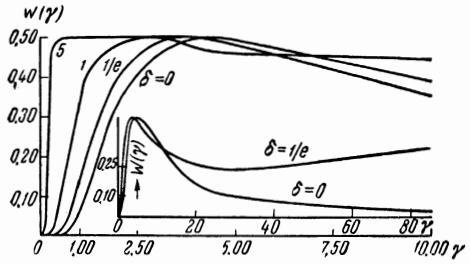

FIG. 3.

The compiled program solves the problem of numerical integration of the system of equations for the two-level model if the electronic matrix elements $H_{11} - H_{22}$ and H_{12} of the interaction are known. By printing-out the values of $|b_2(\tau)|^2$ at the intermediate values of τ we were able to follow directly the positions of the transition regions and their dependences on the collision parameters and on the form of the interaction matrix element. To monitor the correctness of the solution, we used the normalization conservation condition $|b_1|^2 + |b_2|^2 = 1$; in addition, in the case of $\delta = 0$, the numerically obtained solution (that is, the transition probability) coincided within 0.5% with the analytically known result (the Landau-Zener formula).

The cross section of reaction (1) was calculated by numerically averaging the probabilities $W(\rho, v_\infty)$ over all the impact parameters ρ:

$$\sigma(v_\infty) = 2\pi \int_0^{\bar\rho} W(\rho, v_\infty)\rho\, d\rho = \frac{2\pi R_0^2}{\gamma_\infty^2} \int_0^{\gamma_\infty} W(\gamma)\gamma d\gamma,$$
$$\gamma \equiv \gamma_\infty(1 - \rho^2/R_0^2)^{1/2}, \qquad \gamma_\infty \equiv \hbar v_\infty \Delta E/2a^2. \qquad (19)$$

4. DISCUSSION OF RESULTS

Let us compare the calculated transition probability with the deductions of some theoretical papers devoted to charge-exchange reactions. It is expedient to consider separately two ways of improving on the initial Landau-Zener formulation.

1. Account of two transition regions. The problem of calculating the transition probability in this case was discussed by Mordvinov and Firsov[13] and Kozhushner and Sayasov[14]. In the first of these papers, account was taken of both the static and the dynamic coupling of the atomic orbitals of two atoms. Inasmuch as no dynamic coupling is considered in the present paper, direct comparison of the result of the two papers is impossible.

We note, however, that even if this condition is satisfied the plot of the probability (or cross section) vs. v_∞ can have a second maximum—a situation formally analogous to the results of Mordvinov and Firsov. The critical remarks made by Dykhne and Chaplik[15] with regards to the inconsistency of the model employed in [13] cannot pertain to the present case, since the analytic properties of the function ΔE differ greatly here from the properties of the function ΔE of the Landau-Zener model (two branch points).

Kozhushner and Sayasov[14] considered only the static coupling between the atomic functions, and calculated the interference between the transition amplitudes within the framework of the Stueckelberg method, with account of only the branch points closest to the real axis (two branch points for each region; we recall that we are dealing with interference in a single passage through two transition regions; the interference between the incident and scattered waves was not considered in [13] and [14]). The errors due to neglecting in [14] the branch points of ΔE that are far from the real axis were discussed by one of the authors[17]. We note here only that the oscillating addition to the transition probability obtained in [14], as can be seen from the calculation results, is practically nonexistent for the interaction of the type considered here (the crossing of the zeroth-approximation terms in one region and the quasi-crossing in the other).

2. Account of the dependence of the nondiagonal matrix element H_{12} of the interaction between the atomic states on R. This dependence is manifest in the difference between the transition probability W and the quantity W_1 calculated in the Landau-Zener approximation. An approximate account of this dependence is the subject of papers by Dubrovskiĭ[11,12], in which the method of standard equations was used. The formula obtained in these papers for the probability of the charge exchange (1) and proposed for the calculation of the cross section [formula (14) of [11]] differs from the Landau-Zener formula only in the pre-exponential factor $F \equiv [1 + (\alpha v_\infty/\Delta\epsilon)^2]^{-1}$, which leads to a decrease of the probability in the region beyond the maximum as compared with W_1. From the results of the numerical calculation given above and from general considerations it follows that an account of the dependence of H_{12} on R leads to an

increase in the probability on both sides of the maximum. A numerical analysis of the rate at which the transition probability reaches the asymptotic value shows that this result does not change if account is taken of the relatively weak preexponential dependence of H_{12}, or if the expansion of the difference of the zero-order terms near the crossing point is made more precise by adding the nonlinear terms. This difference in the results is connected apparently with the fact that the standard equations used by Dubrovskiĭ correspond to an oversimplified picture of the ion interaction.

Inasmuch as formula (14) of [11] is in better agreement with the majority of the experimental data cited in [12] than the Landau-Zener formula, the discrepancy between the present results and the experimental data in the region of large velocities is greater than in the case of the Landau-Zener formula. It is quite probable that the charge-exchange reactions cited in [12] do not satisfy the limitations of the two-level model, since we are dealing with reactions accompanied by transfer of a p-electron, and consequently all the terms which result from the configurations $(p)^N$ will be close to one another. Exceptions in this respect are the reactions (11/02) and (02/11) for the systems HeLi and AlH. It is to be expected that the assumptions that the width of the transition region is small, $(\Delta R/R_0) \ll 1$, and that the coupling of the two channels in question to all the other channels is weak, $|\sum_{n\neq i} A_{in}/\Delta E_{in}| \ll 1$, are best satisfied near the reaction threshold. In this sense, interest is attached to the reaction (11/20) for the system AlH, for which the cross section near threshold is known [12]. The parameters of this reaction are $\Delta\epsilon = 5.29$ eV, $2a = 1.54$ eV, $\alpha = 1$, and $\delta = 1.45$. It is seen from Fig. 3 that when $\delta \approx 1$ the cross section increases more strongly with increasing γ than in the Landau-Zener approximation. This is in qualitative agreement with the experimental data cited in [12].

The authors thank Professor N. D. Sokolov and M. Ya. Ovchinnikov for a discussion of the work.

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Translated by J. G. Adashko
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