

Quasicrossing of levels and transformation of waves with diffuse states and dissipative factors

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The effect of the finite lifetime of states on nonradiative transitions in diatomic molecules is discussed. It is noted that the finite lifetime leads to an important modification of the transition probability. The latter may become equal to zero for a certain critical value of the lifetime. Some general properties of transition processes, connected with inhomogeneities in plasmas, in diatomic molecules, and in magnetic materials, are analyzed. The dependence of the transition coefficients on the parameters of the system is discussed.

It is well known that systems with inhomogeneous parameters (plasmas with density varying in space, ferromagnets in inhomogeneous magnetic fields, coupled oscillators with time-dependent frequencies, and so on) exhibit a new physical phenomenon, namely, resonant transfer of energy from one state to another in regions where the solutions "cross" (see, for example, [1,2]).

Three types of crossing which differ in their formal and physical criteria are known at present. [2] The first type of crossing of oscillations (states) is characterized by the presence of two points at which the solutions cross in the complex x -plane (x is the coordinate, the variation of which results in a change in the parameters of the system). If one neglects dissipative corrections and other "complex components", the real x axis is transparent to both types of oscillation, and the corresponding dispersion curves do not cross but continuously approach the crossing region. It is important to note that, in this case, the parameters of the system (for example, the electric field and refractive index) do not rise sharply in this region. This distinguishes the first (or sub-barrier) type of crossing from the second anomalous crossing (see [2] for further details). It is also important to note that the transformation coefficient which describes, for example, the transfer of energy from one oscillation to the other, is proportional to $e^{-\delta}$, i.e., it is very sensitive to a change in the parameters of the system on which δ depends.

When dissipative factors are taken into account, the result is a broadening of the resonances of the crossing solutions which, in turn, may have a substantial effect on the transformation process and may, in particular, enhance it. This phenomenon has already been discussed for plasmas. [3,4] Next, it is known that the crossing of the solutions leads to important physical phenomena, not only in classical but also in quantum-mechanical systems (see, for example, [5]). At the same time, we are dealing with an over-barrier type of crossing, so that one would expect that allowance for the complex additions to the energy should appreciably modify the transition probability from one state to another.

The present paper is mainly concerned with the role of "complex components" in quantum-mechanical problems involving the crossing of states.

As an example, consider term crossing in the theory of atomic collisions. The crossing of atomic terms is discussed in many published papers for real values of energy (see, for example, [6,7]). The complex addition to the energy ($E = E_0 + i\Gamma$) arises naturally if we investigate transitions between levels with finite lifetime \hbar/Γ , which leads to the diffuseness of terms. It is then assumed that

the level lifetime is long enough ($\Gamma \ll E_0$) so that transitions from one term to the other do, in fact, succeed in occurring. We next consider certain transition processes connected with inhomogeneities in plasmas, diatomic molecules, and ferro-antiferromagnets. The dependence of transition coefficients on system parameters is then discussed.

1. EFFECT OF TERM DIFFUSENESS ON RADIATIONLESS TRANSITIONS IN DIATOMIC MOLECULES

It is well known [5,6,8] that, in the analysis of transitions between two stationary electronic states $\sigma = 0$ and $\sigma = 1$ with or without change in the orbital angular momentum of the electronic state ($\Lambda_0 \neq \Lambda_1$ and $\Lambda_0 = \Lambda_1$, respectively), the problem can be reduced to a system of two coupled second-order equations:

$$\begin{aligned} U_0'' + \varphi_0 U_0 &= L U_1, & U_1'' + \varphi_1 U_1 &= L U_0, \\ \varphi_0 &= \frac{2M}{\hbar^2} \left[T_0 - E_{\sigma\Lambda_0}(r) - \frac{\hbar^2 J^2}{2M r^2} \right], \end{aligned} \quad (1)$$

where M is the reduced mass of the nuclei, T_0 is the kinetic energy of the nuclei, $E_{\sigma\Lambda_0}$ is the energy of the electrons [measured from $E(r \rightarrow \infty)$], r is the distance between the nuclei, J is the total angular momentum of the system, L is the coupling parameter [for large J , $L = \Lambda(r)J/r^2$], and the primes represent differentiation with respect to r .

Consider the case when either one or both states have a finite lifetime. We shall describe a transition in this system with the aid of (1), obtained from the time-independent Schrödinger equation¹⁾ but with coefficients φ_σ which have imaginary additions Γ_σ due to the finite lifetime of the states.

With regard to the validity of the time-independent Schrödinger equation, we note that the formal situation in the present case is that, in the time-independent formulation, an infinite approach time is required for the collision partner. However, physically, the distances at which boundary conditions corresponding to $r \rightarrow \infty$ can be imposed are of the order of r_0 . Therefore, to describe the transition in the language of the time-independent Schrödinger equation, it is sufficient to satisfy the condition $\hbar^2 \Gamma \tau / M \ll \hbar$, where $\tau \sim r_0/v$ is the characteristic collision time (v is the velocity of the nucleus). We shall assume that this condition is satisfied.

The two equations in (1) reduce to the single fourth-order equation

$$U_0^{(iv)} - 2 \frac{L'}{L} U_0''' + \left[\varphi_0 + \varphi_1 - \frac{L''}{L} - 2 \frac{L'^2}{L^2} \right] U_0'' + \left[2\varphi_0' - \frac{2L'\varphi_0}{L} \right] U_0'$$

$$+ \left[\varphi_0 \varphi_1 - L^2 - 2L' \left(\frac{\varphi_0}{L} \right)' - L'' \frac{\varphi_0}{L} \right] U_0 = 0, \quad (2)$$

$$U_1 = L^{-1} [U_0'' + \varphi_0 U_0].$$

In accordance with the WKB method, the general solution of (2) will be written in the form

$$U_0 \approx \left[\left(c_+ \exp \left\{ i \int v_0 dr \right\} + c_- \exp \left\{ -i \int v_0 dr \right\} \right) \exp \left(\frac{1}{2} \left(- \int \frac{dv_0}{v_0} + \int \frac{dt}{(1+t^2)^{1/2}} \right) \right) + \left(d_+ \exp \left\{ i \int v_1 dr \right\} + d_- \exp \left\{ -i \int v_1 dr \right\} \right) \right] \exp \left(- \frac{1}{2} \int \frac{t dt}{1+t^2} \right); \quad (3)$$

$$v_{0,1} = \varphi \pm (\Psi^2 + L^2)^{1/2}, \quad \varphi = 1/2(\varphi_0 + \varphi_1), \quad \Psi = 1/2(\varphi_0 - \varphi_1),$$

$$t = \Psi/L,$$

where c_{\pm} and d_{\pm} are coefficients.

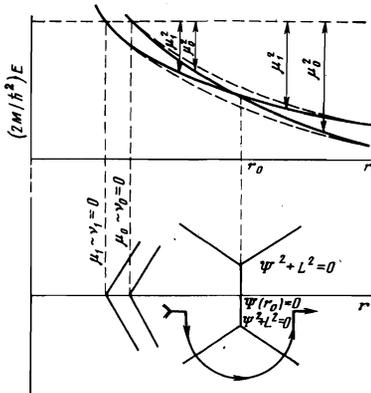
This solution is valid everywhere except for the neighborhoods of the points $\nu_0 = 0$, $\nu_1 = 0$, $\nu_0 = \nu_1$. We shall be interested in the $\nu_0 = \nu_1$ crossing, illustrated in the figure, for which $\Lambda_0 \neq \Lambda_1$. At points $\Psi^2 + L^2 = 0$, which are located symmetrically relative to the r axis, we have $\nu_0 = \nu_1$; three Stokes lines emerge from each point, of which only one cuts the r axis and joins these points with one another. The solution on the left of the crossing points is obtained from (3), in which case

$c_{\pm} = c e^{\pm 1/4 i \pi}$ and $d_{\pm} = d e^{\pm 1/4 i \pi}$, and the limits of integration are chosen so that the lower limits correspond to values of r for which $\nu_{0,1} = 0$ and the upper limits are r_0 . To obtain the solutions to the right of the crossing point r_0 , we must bypass the point $\nu_0 = \nu_1$ in the complex region, as shown in the figure. The coefficients undergo discontinuities when the Stokes lines are intersected. The parameters which describe these discontinuities can be found unambiguously to within a phase factor for a unitary transition matrix.^[9] This matrix describes transitions in dissipative systems. It is shown in the Appendix that the unitarity conditions used for dissipative systems for the transitions matrix are possible when the parameter Γ/φ is small. It will be shown later that the transition coefficient is determined by the parameter Γ/L which may be close to unity for $\Gamma/\varphi \ll 1$ since $L \ll \varphi$ always and, therefore, we shall assume that $\Gamma/\varphi \ll 1$ when we consider the transition.

Consider Ψ in the neighborhood of r_0 . Since we are assuming that the energies of the electron terms have imaginary additions, we may write

$$\Psi = |F_1 - F_0| (r - r_0) + i\Gamma, \quad F_{0,1} = \frac{1}{2} \frac{d\varphi_{0,1}}{dr} \Big|_{r=r_0},$$

where Γ may be either positive or negative. In the same neighborhood, L may be assumed to be constant. Pro-



ceeding by analogy with,^[6] and bypassing the point $\nu_0 = \nu_1$, we obtain the solution asymptotic in r to the right of r_0 :

$$U_0 = \left(\frac{2}{v_0} \right)^{1/2} \left[(ce^{-\delta} + d(1 - e^{-2\delta})^{1/2} e^{-i\tau}) \exp \left(i \int_{v_0=0}^r v_0 dr - i \frac{\pi}{4} \right) + (ce^{-\delta} + d(1 - e^{-2\delta})^{1/2} e^{i\tau}) \exp \left(i \int_{v_0=0}^r v_0 dr + i \frac{\pi}{4} \right) \right],$$

$$U_1 = - \left(\frac{2}{v_1} \right)^{1/2} \left[(-c(1 - e^{-2\delta})^{1/2} e^{i\tau} + de^{-\delta}) \exp \left(i \int_{v_1=0}^r v_1 dr - i \frac{\pi}{4} \right) + (-c(1 - e^{-2\delta})^{1/2} e^{-i\tau} + de^{-\delta}) \exp \left(-i \int_{v_1=0}^r v_1 dr + i \frac{\pi}{4} \right) \right], \quad (4)$$

$$\delta_0 = \frac{L^2 \pi}{2|F_1 - F_0| (v_1 + v_0)}, \quad \tau = \int_{v_0=0}^r v_0 dr - \int_{v_1=0}^r v_1 dr,$$

$$\delta = \delta_0 \left\{ 1 - \frac{2|\Gamma|}{\pi L} \left(1 - \frac{\Gamma^2}{L^2} \right)^{1/2} - \frac{2}{\pi} \arcsin \frac{|\Gamma|}{L} \right\}, \quad \Gamma \leq L, \quad \delta = 0, \quad \Gamma > L.$$

We shall be interested in the transition of the molecule from one electronic state to another when the crossing point r_0 is traversed twice. Suppose that the molecule is initially in the $\sigma = 0$ state, and that its final state can be either $\sigma = 0$ or $\sigma = 1$. The boundary conditions at infinity for the nuclear wave function, which correspond to this situation in the WKB approximation, are

$$\lim_{r \rightarrow \infty} U_0 = \left(\frac{2}{\pi \eta_0} \right)^{1/2} \left[\left(\eta_0 + \frac{1}{2} \right) e^{i\beta} \exp \left(i \int_{v_0=0}^r v_0 dr - i \frac{\pi}{4} \right) + \frac{1}{2} e^{-i\beta} \exp \left(-i \int_{v_0=0}^r v_0 dr + i \frac{\pi}{4} \right) \right], \quad (5)$$

$$\lim_{r \rightarrow \infty} U_1 = \left(\frac{2}{\pi \eta_1} \right)^{1/2} \left[\eta_1 e^{-i\gamma} \exp \left(i \int_{v_1=0}^r v_1 dr - i \frac{\pi}{4} \right) \right],$$

$$\beta = \lim_{r \rightarrow \infty} \left(\int_{v_0=0}^r v_0 dr - \int_{\mu_0=0}^r \mu_0 dr \right), \quad \gamma = \lim_{r \rightarrow \infty} \left(- \int_{v_1=0}^r v_1 dr + \int_{\mu_1=0}^r \mu_1 dr \right).$$

If we satisfy these boundary conditions, we obtain a set of equations for the constants η_0 , η_1 , c , d , which yield

$$c = \frac{1}{2} \pi^{-1/2} e^{-\delta - i\beta}, \quad d = \frac{1}{2} \pi^{-1/2} (1 - e^{-2\delta})^{1/2} e^{-i(\beta + \tau)}$$

$$\eta_0 = -i [e^{-i\beta} + (1 - e^{-2\delta}) e^{-i(2\beta + \tau)} \sin \tau],$$

$$\eta_1 = i (1 - e^{-2\delta})^{1/2} e^{-i(\beta + \tau) - \delta} \sin \tau.$$

The flux of U_0 is then proportional to

$$(2/i) U_0 \bar{U}_0 = \text{Re} (\eta_0 + \eta_0 \bar{\eta}_0) = -(1 - e^{-2\delta}) e^{-2\delta} \sin^2 \tau.$$

The flux of U_1 is proportional to

$$2\pi^{-1} U_1 \bar{U}_1 = \eta_1 \bar{\eta}_1 = (1 - e^{-2\delta}) e^{-2\delta} \sin^2 \tau. \quad (6)$$

It is important to note that this transition corresponds to two transits through r_0 . The first of the factors, $1 - e^{-2\delta}$, is connected with the transition probability from one term to the other when the point r_0 is traversed, and the second, $e^{-2\delta}$, is connected with the probability of remaining on the same term. When $\Gamma = 0$, we obtain the result reported by Stueckelberg,^[6] in which case the maximum value $|\eta_1^0|^2 = 1/4 \sin^2 \tau$ is reached for $\delta_0 = 1/2 \ln 2$ (where $|\eta_1^0|^2$ is the transition probability without allowing for the term diffuseness). Since $\Psi = 1/2(\varphi_0 - \varphi_1)$, the imaginary addition Γ is determined by the difference between the imaginary additions to φ_0 and φ_1 , which correspond to the additions $\hbar^2 \Gamma_{0,1}/M$ to the term energy, which in turn are determined by the finite lifetime of the state. Suppose that the energy of the first term has the imaginary addition $\hbar^2 \Gamma/M$, i.e., this state has a finite

lifetime. At the critical value of the lifetime, when $|\Gamma| = L$, the point $\nu_0^2 = \nu_1^2$ lies on the real axis of r . In this case, $|\eta_1|^2 = 0$, i.e., the flux of U_1 is zero, which corresponds to zero transition probability. It is clear from the expression for δ that, for large δ_0 , when $|\eta_0|^2$ is close to zero, the probability may be a maximum because of the finite lifetime of the state.

2. SOME GENERAL PROPERTIES OF TRANSITION PROCESSES CONNECTED WITH INHOMOGENEITY IN PLASMAS, DIATOMIC MOLECULES, AND FERRO-ANTIFERROMAGNETS

In this section, we consider the transformation of spin waves into acoustic waves near the ferroacoustic resonance, the transformation of the extraordinary wave into the ordinary wave near the total reflection point for an ordinary wave, and transitions between terms in a diatomic molecule near their crossing point with a view to establishing the general features of these phenomena. We have to consider the coupled second-order equations

$$U_{0,i}'' + (\varphi_{0,i} + aL^2)U_{0,i} = \left(b \frac{dL}{dz} + cL \right) U_{1,i} + d_{0,i}LU_{1,i},$$

in which the coupling is produced by the quantities under investigation themselves (diatomic molecule, $c = 1$, $a = b = d_{0,1} = 0$),^[6] or their derivatives (ferro-antiferromagnet; $d_{0,1} = \pm 1$, $a = b = c = 0$ ^[10,11]), or in mixed form (plasmas, $a = 1$, $b = -1$, $c = 0$, $d_{0,1} = -2$ ^[3,4]). The coupled equations have two wave-number branches for normal oscillations. The above processes correspond either to transition between branches of normal oscillations (plasmas), or to the displacement along one of the branches (ferro-antiferromagnet), or to the sum of such transitions (diatomic molecule). The form of the wave-number branches for normal oscillations near the transition point is illustrated in the figure.

The distance between the branches is determined by the coupling parameters L . The plasma coupling parameter is, in this case, a function of the damping Γ :

$$L = -i \frac{dv}{dz} \frac{\Gamma_{cr}}{(v-i(\Gamma_{cr}+\Gamma)-1)(v+i(\Gamma_{cr}-\Gamma)-1)},$$

where $v = 4\pi e^2 N(z)/m\omega^2$, $N(z)$ is the plasma density, $\Gamma_{cr} = eH_0 \sin^2 \theta / mc\omega |\cos \theta|$, and θ is the angle between the wave vector and the magnetic field H_0 . In the diatomic molecule, L is independent of Γ , and in the ferro-antiferromagnet $L = 2\gamma^2 M_0^2 / \rho c_t^2 \alpha$, where γ is the magnetostriction constant, c_t is the velocity of transverse sound waves, M_0 is the magnetic moment, and α is the elastic constant.

Because of the presence of complex additions Γ which are connected with collisions (plasmas), relaxation (ferro-antiferromagnets), and finite lifetime of the level (diatomic molecules), the separation between the normal oscillation curves may alter when the curves merge for certain critical values Γ_{cr} . The presence of the complex additions increases the probability of transition between the normal oscillation branches, thus affecting the probability of the physical transition under consideration. The transformation coefficient which describes the transformation of the extraordinary wave into the ordinary wave in inhomogeneous plasmas in the presence of collisions is obtained in^[3,4]:

$$\delta = \delta_0 \left(1 - \frac{\Gamma}{\Gamma_{cr}} \right)^{1/2}, \quad K = e^{(-\delta)}, \quad \delta_0 = \pi k_0 \Gamma_{cr} / \frac{1}{\beta_0} \left(\frac{dH}{dz} \right)_{z=z_p},$$

where k_0 is the wave vector and β_0 is a numerical factor. The coefficient K is a measure of the fraction of the

incident-wave amplitude which, having reached the transformation point, is transformed into another type of wave (without taking into account the reduction in amplitude due to collisions up to the point of transformation).

Schlömann and Joseph^[10] have considered the transformation of a spin wave into an acoustic wave in a ferromagnet placed in an inhomogeneous magnetic field to ensure ferroacoustic resonance, without taking into account spin-wave damping. Bar'yakhtar et al.^[11] have obtained an expression for the transformation coefficient describing the change in the spin-wave amplitude between the antiferromagnetic and the ferroacoustic resonances (see, for example,^[12]). To obtain the transformation coefficient for the spin wave generated at the antiferromagnetic resonance point into an acoustic wave, the coefficient given in^[11] must be multiplied by the coefficient representing the transformation of these waves at the ferroacoustic resonance point. In the absence of damping, this coefficient is given by^[10]

$$K = 1 - \exp(-\delta_0)$$

and for the usually encountered magnetic field inhomogeneities, it approaches unity, so that the total transformation coefficient is the same as in^[11].

On the other hand, in the presence of spin-wave damping approaching the critical value Γ_{cr} , the argument of the exponential acquires an additional factor close to zero, which ensures that the coefficient describing the transformation of the spin wave into the acoustic wave is small, so that the total transformation coefficient is small. Since the equations for the wave numbers of atomic molecule are similar, calculations of the factor in the argument of the exponential, δ , which depends on the damping coefficient κ , are analogous to the calculations performed in the preceding section and will therefore not be reproduced here. The final result is

$$\delta = \delta_0 \left[1 - \frac{2\kappa}{\pi \Gamma_{cr}} \left(1 - \left(\frac{\kappa}{\Gamma_{cr}} \right)^2 \right)^{1/2} - \frac{2}{\pi} \arcsin \left(\frac{\kappa}{\Gamma_{cr}} \right) \right],$$

$$\delta_0 = 2M_0 \pi k_t L^2 / \left(\frac{dH}{dz} \right)_{z=z_p}, \quad \Gamma_{cr} = \frac{2k_t L}{(k_t^2 - L^2)^{1/2}},$$

where k_t is the wave number of the transverse acoustic wave. Consequently, the presence of the complex additions may enhance physical transitions connected with the transition between the branches of normal oscillations (plasma and diatomic molecule) and may weaken transitions connected with the transition along one of the branches (ferro-antiferromagnet).

Finally, we note some general effects. The form of the expression for the wave numbers of normal oscillations completely determines the dependence of the argument of the exponential on the ratio Γ/Γ_{cr} . If the coupling between the equations is produced by the quantities under consideration themselves, then δ is inversely proportional to the wave number. On the other hand, if the coupling is through the derivatives, then it is proportional to the wave number. The numerator of the argument of the exponential always includes the derivative of the quantity defining the "angle" between normal oscillation branches in the particular transition.

APPENDIX

It is well-known that the unitary transformation matrix conserves the vector norm and, conversely, if the vector norm is conserved, the transformation matrix is unitary. Consider the two coupled equations with complex coefficients:

$$U_0'' + \varphi_0(r)U_0 = L(r)U_1, \quad U_1'' + \varphi_1(r)U_1 = L(r)U_0; \quad (A.1)$$

$$\varphi_{0,1} = \varphi_{0,1} - i\Gamma_{0,1}.$$

Near the point $\Omega_1 = \Omega_2$ we then have

$$\Omega_{1,2} = \frac{1}{2} \{ \varphi_0 + \varphi_1 - i(\Gamma_0 + \Gamma_1) \pm [(\varphi_0 - \varphi_1 - i(\Gamma_0 - \Gamma_1))^2 + 4L^2]^{1/2} \},$$

and from (A.1) we have

$$i \frac{\Gamma_0 - \Gamma_1 + \Phi \sin \chi}{\Pi_0^2} \left\{ \exp \left(2 \int_{r_1}^r \Pi_0^2 \sin \rho_0 dr \right) |A|^2 \right.$$

$$- \frac{\Pi_0^2}{\Pi_1^2} \exp \left(2 \int_{r_1}^r \Pi_1^2 \sin \rho_1 dr \right) |B|^2 \left. + 2i \left[\cos \rho_0 \exp \left(2 \int_{r_1}^r \Pi_0^2 \sin \rho_0 dr \right) \frac{d}{dr} |A|^2 \right. \right.$$

$$\left. \left. + \cos \rho_1 \exp \left(2 \int_{r_1}^r \Pi_1^2 \sin \rho_1 dr \right) \frac{d}{dr} |B|^2 \right] = 0,$$

where A and B are the amplitudes in the WKB approximation corresponding to the frequencies Ω_1 and Ω_2 :

$$\Pi_{0,1}^2 = (1/4) \{ (\varphi_0 + \varphi_1 \pm \Phi \cos \chi)^2 + (\Gamma_0 + \Gamma_1 \pm \Phi \sin \chi)^2 \}^{1/2},$$

$$\rho_{0,1} = \frac{1}{2} \operatorname{arctg} \left(\frac{\Gamma_0 + \Gamma_1 \pm \Phi \sin \chi}{\varphi_0 + \varphi_1 \pm \Phi \cos \chi} \right),$$

$$\Phi = \{ [(\varphi_0 - \varphi_1)^2 + 4L^2 - (\Gamma_0 - \Gamma_1)^2] + 4(\Gamma_0 - \Gamma_1)^2(\varphi_0 - \varphi_1)^2 \}^{1/2},$$

$$\chi = \frac{1}{2} \operatorname{arctg} \left(\frac{2(\Gamma_0 - \Gamma_1)(\varphi_0 - \varphi_1)^2}{(\varphi_0 - \varphi_1)^2 + 4L^2 - (\Gamma_0 - \Gamma_1)^2} \right).$$

When $\Gamma_0 = \Gamma_1 = 0$, we have the usual conservation law (see, for example, [9])

$$\frac{d}{dr} (|A|^2 + |B|^2) = 0, \quad \text{i. e. } |A|^2 + |B|^2 = \text{const.}$$

Therefore, the vector norm is an invariant quantity near r_0 , where $\Omega_1 = \Omega_2$. Consequently, the transition matrix describing the matching of asymptotic solutions is unitary.

When $\Gamma/\varphi \ll 1$, the conservation law is satisfied approximately and, therefore, the matrix is approximately unitary. The admissible error is then determined by the ratio Γ/φ . The transition matrix corresponding to the case considered in the first section then has a form analogous to that given in [9]:

$$M = \begin{pmatrix} ie^{i\Phi}(1-e^{-2\delta})^{1/2} & e^{-\delta} \\ e^{-\delta} & ie^{-i\Phi}(1-e^{-2\delta})^{1/2} \end{pmatrix}.$$

The difference lies only in the definition of δ :

$$\delta = \frac{i}{2} \int_C (\Omega_1 - \Omega_2) dr$$

where the contour C joins segments of the real axis and surrounds the point $\Omega_1 = \Omega_2$ nearest to this axis.

We are indebted to N. S. Erokhin and V. V. Gann for useful discussions.

¹An example of the application of the time-independent formulation of the problem to a system with a finite lifetime is the problem of resonance on quasiscrete levels, which is discussed by Landau and Lifshitz [5].

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