Landau-Zener transition to a decaying level

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Exact analytical solutions for the problem of term crossing are obtained for decaying levels with the help of the Laplace contour-integral method. For the case of fast passage through resonance and for the case of slow decay, simple asymptotic expressions are found from the exact solution in terms of Whittaker functions.

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Consider a two-level quantum particle in an intense laser field. Let the detuning between the laser and the transition frequency depend linearly on time. What is the probability of finding the particle in the upper state after a single passage through the resonance? We immediately recognize here the well-known Landau-Zener problem [1, 2], which has been the subject of intensive study during the past 60 years. This problem appears in atomic collisions and physical chemistry when nonadiabatic transitions are of importance [3–6], in semiconductors [7], in particle and nuclear physics [8], in atomic interferometry [9, 10], and in the interaction of atoms and molecules with laser fields [11–14]. Moreover, a particle with the spin $\frac{1}{2}$ moving in an inhomogeneous magnetic field [15], and regular above-barrier scattering, are described by the Schrödinger equation of the same type and allow the same semiclassical approach [16–21]. One can also apply this model to describe resonance-enhanced multiphoton ionization (REMPI) of atoms by a laser in the case when the laser field has a fixed frequency but the intensity is time dependent [22]. Time-dependent detuning results then from the second-order Stark shift of the atomic levels.

For an atom initially in the lower level, the Landau-Zener model suggests the expression for the population of the upper level after a single passage through resonance,

$$\rho_{22} = 1-\rho_{11} = 1-e^{-2\pi V^2/\alpha}. \tag{1}$$

Here α is the slope of the time-dependent detuning $\Delta = \alpha t$, and V denotes the interaction amplitude divided by $\hbar$. But what happens if the upper level can decay? The present paper answers this question for two types of decay: (i) irreversible decay of the upper level to a continuum, and (ii) longitudinal relaxation of the upper level to the lower state.

These problems become important in the context of laser-induced level crossing [19, 23, 24, 26, 11–13], that is, crossing of dressed states of atoms or quasienergy terms of molecules. Photoinduced decomposition of particles and spontaneous emission of photons are the physical processes responsible for the decay in the above cases (i) and (ii), respectively.

The main tool of our analysis is the Laplace contour integral method [1]. It enables us to obtain exact analytical solutions of the problems (i) and (ii) in terms of integrals that in fact correspond to special functions: the parabolic cylinder function in case (i) and the Whittaker function in case (ii). We evaluate the integrals using the stationary phase method and get the asymptotics of these functions.

We consider case (i) first. The Schrödinger equation for the probability amplitudes ψ_1 and ψ_2 for the lower and upper level reads

$$i\dot{\psi}_1 = V\psi_2, \tag{2}$$

$$i\dot{\psi}_2 = \alpha t\psi_2 - i\gamma\psi_2 + V\psi_1. \tag{3}$$

Here $2V$ is the Rabi frequency and γ is the relaxation rate. The initial condition is

$$\psi_1(\infty) = 1. \tag{4}$$

Substitution of Eq. (2) into Eq. (3) yields

$$\ddot{\psi}_1 + i\alpha t\dot{\psi}_1 - \gamma\dot{\psi}_1 + V^2\psi_1 = 0. \tag{5}$$

The Laplace contour-integral method suggests a solution of Eq. (5) in the form

$$\psi_1 = A\int_C \exp\left\{\frac{-i\tau^2}{2\alpha} + i\frac{\gamma}{\alpha}\tau + \tau t\right\}\frac{d\tau}{\tau^{1+iV^2/\alpha}}, \tag{6}$$

where A is a normalization constant. The contour C, which allows us to satisfy in the simplest way the initial condition, Eq. (4), comes in from infinity along the direction $(1-i)$ and returns back to infinity after the circumvention around the point $\tau = 0$. Equation (6) resembles the integral representation of the parabolic cylinder functions $D_p(z)$ [25], and indeed, the exact time-dependent solution of Eqs. (2) and (3) reads

$$\psi_1 = A' e^{\frac{-i(i\gamma+\alpha t)^2}{4\alpha}} D_{\frac{iV^2}{\alpha}}\left[-e^{\frac{-i\pi}{4}}\frac{(i\gamma+\alpha t)}{\sqrt{\alpha}}\right]. \tag{7}$$

Note that this result is the analytical continuation of the exact solution of the Landau-Zener problem for nondecaying levels [8, 18] with $t \to t + i\gamma/\alpha$. We find the constant A in Eq. (6) from the initial condition Eq. (4) and the asymptotic expression of the integral Eq. (6): At the extreme $t = -t_0 \to -\infty$ we take $\tau \sim 1/t_0$ in the integrant, neglect the first and the second term in the exponent, rotate the contour C by the angle $\pi/4$, and thereby reduce the integral to the integral representation of the Γ function. Finally we get

$$A = \frac{\exp(-\frac{iV^2}{\alpha}\ln t_0 - \frac{\pi V^2}{4\alpha})}{\Gamma(\frac{iV^2}{\alpha})}. \tag{8}$$

We now ask what is the population $\rho_{22} = |\psi_2|^2$ of the upper level as $t \to \infty$? Due to the decay of the level it is apparently zero. Now, what is the population $\rho_{11} \equiv |\psi_1|^2$ of the lower level in this limit? Expression (6) provides the answer to this question when we evaluate the integral using the method of stationary phase. There are two stationary phase points: One at $\tau = -\gamma + i\alpha t$ that brings in a contribution of the order of $\frac{1}{t}$ which is negligible for large t, and another that is close to $\tau = 0$. Hence we can neglect the τ^2 term in the exponent. After a rotation of the contour by $3\pi/4$, the integral reduces to the integral representation of the Γ function and yields

$$\psi_1 = \exp\left(-\frac{\pi V^2}{\alpha} - \frac{iV^2}{\alpha}\ln|tt_0|\right), \tag{9}$$

that is,

$$\rho_{11} = \exp\left(-\frac{2\pi V^2}{\alpha}\right). \tag{10}$$

The population of the ground state is identical to that given by the Landau-Zener formula, Eq. (1), which does not take into account the relaxation process. In other words, irreversible decay of the upper level does not affect the probability of a Landau-Zener transition.

Now let us consider a Landau-Zener transition in a two-level system with longitudinal relaxation, that is, case (ii). The population of the upper level does not leave the system after the relaxation but returns to the ground state. This is a non-Hamiltonian system, and hence for its description we have to employ the density-matrix equation or the equivalent set of Bloch equations [27]. The Bloch equations for polarization $\mathcal{P} = \rho_{12} + \rho_{21}$, dispersion $\mathcal{Q} = \rho_{12} - \rho_{21}$, and population of the upper level $\rho_{22} \equiv \frac{1}{2}\mathcal{Z}$ reads

$$\begin{aligned} \dot{\mathcal{Z}} &= 2V\mathcal{P} - \gamma\mathcal{Z}, \\ \dot{\mathcal{P}} &= -2V\mathcal{Z} + \alpha t\mathcal{Q} - \gamma\mathcal{P} + 2V, \\ \dot{\mathcal{Q}} &= -\alpha t\mathcal{P} - \gamma\mathcal{Q}. \end{aligned} \tag{11}$$

Here we have assumed that the constant γ is identical for the transverse and longitudinal relaxation. The initial conditions are

$$\mathcal{Z}(-\infty) = \mathcal{P}(-\infty) = \mathcal{Q}(-\infty) = 0. \tag{12}$$

At large positive times the two-level system is out of resonance, and hence the population returns to the lower level as a result of the longitudinal relaxation. This gives the boundary condition

$$\mathcal{Z}(\infty) = \mathcal{P}(\infty) = \mathcal{Q}(\infty) = 0, \tag{13}$$

which, together with Eqs. (12), allows us to perform a Fourier transformation of Eq. (11),

$$\begin{aligned} i\omega\mathcal{Z} &= 2V\mathcal{P} - \gamma\mathcal{Z}, \\ i\omega\mathcal{P} &= -2V\mathcal{Z} + i\alpha\frac{\partial}{\partial\omega}\mathcal{Q} - \gamma\mathcal{P} + 4\pi V\delta(\omega), \\ i\omega\mathcal{Q} &= -i\alpha\frac{\partial}{\partial\omega}\mathcal{P} - \gamma\mathcal{Q}. \end{aligned} \tag{14}$$

Now we express $\mathcal{Z}$ and $\mathcal{Q}$ in terms of $\mathcal{P}$ with the help of the first and the third equation of Eqs. (14) and substitute them into the second equation of this set. This yields

$$\frac{\alpha^2}{\omega - i\gamma}\frac{\partial}{\partial\omega}\frac{1}{\omega - i\gamma}\frac{\partial}{\partial\omega}\mathcal{P} + \mathcal{P} - \frac{4V^2}{(\omega - i\gamma)^2}\mathcal{P} = 4\pi\frac{V}{\gamma}\delta(\omega). \tag{15}$$

The Dirac δ function at the right-hand side of the equation implies that at the point $\omega = 0$ the Fourier transform of polarization $\mathcal{P}(\omega)$ has a discontinuity in the first derivative. When we integrate Eq. (15) over a small vicinity ϵ of $\omega = 0$ we find

$$\left.\frac{\partial\mathcal{P}}{\partial\omega}\right|_{\omega=0+\epsilon} - \left.\frac{\partial\mathcal{P}}{\partial\omega}\right|_{\omega=0-\epsilon} = -4\pi\frac{\gamma V}{\alpha^2}. \tag{16}$$

Thus, we have to find the solution of the homogeneous equation (15) that satisfies the boundary condition, Eq. (16), and corresponds to a real $\mathcal{P}(t)$, that is,

$$\mathcal{P}(\omega) = \mathcal{P}^*(-\omega). \tag{17}$$

We now may obtain the exact analytical solution of Eq. (15) in terms of Whittaker functions. In order to simplify the notations, we scale all frequencies with the Rabi frequency, that is, we set $2V = 1$, $\gamma = \gamma/(2V)$, $\alpha = \alpha/(2V)^2$, $\omega = \omega/(2V)$, and introduce the new variable $x = \frac{(i\omega+\gamma)^2}{2\alpha}$. Then we come to

$$x\frac{\partial^2}{\partial x^2}\mathcal{P} + \frac{1}{2\alpha}\mathcal{P} + x\mathcal{P} = 0. \tag{18}$$

The Laplace contour-integral method gives the solution of this equation,

$$\mathcal{P} = \frac{A'}{2i\alpha}\int_{C_W}\frac{\exp(\tau x)}{(\tau + i)^{1-i/4\alpha}(\tau - i)^{1+i/4\alpha}}d\tau. \tag{19}$$

The contour C_W comes in from $\tau = i\infty$, goes around the point $\tau = i$, and goes back along the direction $\tau = i\infty$. This integration path ensures convergence of the integral for $\omega > 0$. The integral representation for $\omega < 0$ follows from the condition Eq. (17) [29].

The substitution $\tau = i + i2z$ transforms the integral, Eq. (19), to the integral representation of the Whittaker function [28],

$$u_1(\omega) = W_{i/4\alpha,-1/2}\left(-i\frac{(i\omega + \gamma)^2}{\alpha}\right). \tag{20}$$

Here, we have made use of the explicit expression for x. Hence, the Whittaker function $u_1(\omega)$ is a solution of Eq. (18) for $\omega > 0$, that is, $\mathcal{P}(\omega)|_{\omega>0} = Au_1(\omega)$. The other linearly independent solution of Eq. (15),

$$\mathcal{P}(\omega) = A^*u_2(\omega) = A^*W_{-i/4\alpha,-1/2}\left(i\frac{(i\omega + \gamma)^2}{\alpha}\right) \tag{21}$$

accounts for the case $\omega < 0$.

Now, we can find the proportionality constant A with the help of the condition given in Eq. (17) at $\omega = 0$ and the condition Eq. (16). We come then to the set of equations

$$A u_1(0) - A^* u_2(0) = 0,$$
$$A \frac{\partial u_1}{\partial \omega}\Big|_{\omega=0} - A^* \frac{\partial u_2}{\partial \omega}\Big|_{\omega=0} = -2\pi \frac{\gamma}{\alpha^2}. \tag{22}$$

The determinant D of this set is proportional to the Wronskian $W(u_1;u_2) \equiv (u_1 \frac{\partial}{\partial x} u_2 - u_2 \frac{\partial}{\partial x} u_1)$ of the Whittaker functions [30] and reads

$$D = \frac{2\gamma}{\alpha} W(u_1;u_2) = \frac{2\gamma}{\alpha} \exp\left(\frac{\pi}{4\alpha}\right). \tag{23}$$

The factor $\frac{2\gamma}{\alpha}$ results from the derivative of the arguments $\frac{\partial x}{\partial \omega}$. Solving Eq. (22) we obtain the constant A and find for the polarization

$$\mathcal{P}(\omega)|_{\omega>0} = \pi \frac{u_2(0) u_1(\omega)}{\alpha} \exp\left(-\frac{\pi}{4\alpha}\right). \tag{24}$$

Now, we are able to calculate the number of photons $I = \gamma \int \rho_{22}(t) dt = \gamma \rho_{22}(\omega = 0)$ emitted per atom after the passage through the resonance. Since, according to Eq. (14), we have $\rho_{22}(\omega=0) = \frac{1}{2}\mathcal{Z}(\omega=0) = (2\gamma)^{-1}\mathcal{P}(\omega=0)$ we find from Eqs. (20), (21), and (24) for $\omega = 0$

$$I = 2\pi \frac{V^2}{\alpha} e^{\frac{-\pi V^2}{\alpha}} \left| W_{\frac{iV^2}{\alpha},-\frac{1}{2}}\left(-\frac{i\gamma^2}{\alpha}\right)\right|^2. \tag{25}$$

Here we have used the original dimensional variables. This expression is the final result of the exact analytical consideration.

In Fig. 1, we show the dependence of the yield I on the Landau-Zener parameter $\frac{V^2}{\alpha}$ and on the quenching parameter $\frac{\gamma^2}{\alpha}$. At $\gamma = 0$, the yield is given by the regular

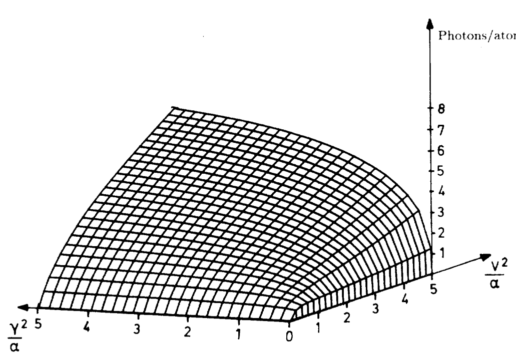

FIG. 1. Expectation value of the number of photons emitted per atom given by Eq. (25) as a function of Landau-Zener parameter V^2/α and quenching parameter γ^2/α. The Whittaker function was calculated numerically with the help of the integral representation Eq. (26) after the substitution $t = e^x$. [Numerical solution of the set of Bloch equations Eq. (11) gives the same result.]

Landau-Zener dependence Eq. (1). It changes with the increase of the quenching parameter γ^2/α, as the number of spontaneously emitted photons per atom increases with increasing relaxation rate. This implies that laser photons get transformed into the spontaneously emitted photons during passage through the resonance.

We now consider the asymptotic behavior of Eq. (25). We start with the case of slow passage through the resonance, that is, $\alpha \to 0$. We use the integral representation [30] of the Whittaker function and find with the help of the method of stationary phase the following expression:

$$W_{\frac{iV^2}{4\alpha},-\frac{1}{2}}\left(-\frac{i\gamma^2}{\alpha}\right)$$
$$= \frac{e^{i\gamma^2/2\alpha}}{\Gamma(-\frac{iV^2}{\alpha})} \int_0^\infty \frac{\exp\{i\frac{\gamma^2}{\alpha}t - i\frac{V^2}{\alpha}\ln\frac{t}{1+t}\}}{t(t+1)} dt$$
$$\simeq \frac{e^{i\Phi}}{\Gamma(-i\frac{V^2}{\alpha})} \frac{\sqrt{\pi\alpha}}{[1+(\frac{2V}{\gamma})^2]^{1/4} V}, \tag{26}$$

where Φ is a phase factor that does not effect the final result, Eq. (25). Here, we have taken into account that in the limit $\alpha \to 0$ the only stationary point on the integration path is $t = -\frac{1}{2} + \frac{1}{2}\sqrt{1 + \frac{4V^2}{\gamma^2}}$. We also have assumed $V^2\gamma^2/\alpha^2 \gg 1$, allowing us to neglect derivatives higher than second order at the stationary phase point and thus satisfy the requirement of the stationary phase method. Substitution of Eq. (26) into Eq. (25) results in

$$I = 2\pi \frac{\gamma V^2}{\alpha} \frac{1 - \exp\{\frac{-2\pi V^2}{\alpha}\}}{(\gamma^2 + 4V^2)^{1/2}}, \tag{27}$$

where we have made use of the formula $|\Gamma(iy)|^2 = \pi(y \sinh \pi y)^{-1}$. The exponential term reminiscent of the Landau-Zener expression arises from the Γ function and not from the stationary phase analysis of Eq. (26). However, in order to be consistent within the order of approximation it should be neglected. Note that this result also follows from the rate equation [31]. The limit of weak interaction, $V^2/\alpha \to 0$, follows from the condition $|W_{0,-1/2}(x)| = |e^{ix}| = 1$, and Eq. (25) simplifies to $I \simeq 2\pi V^2/\alpha$. This implies that the asymptotic formula Eq. (27) is also valid for weak interactions.

We now consider the limit of slow decay, $\frac{\gamma^2}{\alpha} \ll 1$, with the help of the asymptotic expression for Whittaker function [30],

$$\lim_{z\to 0} W_{ia,-\frac{1}{2}}(z) = \frac{1}{\Gamma(1+ia)} = \frac{1}{ia\Gamma(ia)}, \tag{28}$$

which, after substitution into Eq. (25), results in

$$I = 1 - \exp\left(-\frac{2\pi V^2}{\alpha}\right). \tag{29}$$

We hence recover the probability of the Landau-Zener transition, Eq. (1), which indeed is identical to the probability of emission of a spontaneous photon in the case of a small decay rate. We note that a simple expression for the limit $V^2\gamma^2/\alpha^2 \ll 1$ is difficult to obtain due to the nonanalytical behavior of the Whittaker function at small values of the argument, shown in Fig. 1.

We conclude by summarizing the main results of the present paper that follow from exact analytical solutions of the Landau-Zener problem obtained for the case of relaxation of the upper level to a continuum, and for the case of longitudinal relaxation of the upper level to the lower state. Rabi oscillations of population between the upper and the lower states, which take place while the levels are in resonance [21], do not manifest themselves in any oscillations of the regular Landau-Zener transition probability. At first sight, one might think that the relaxation would bring them to light since it interrupts the population oscillations in the middle of the Rabi cycle, that is, before the passage through the resonance is complete. However, they remain hidden even in the case when relaxation processes are taken into account: The irreversible decay of the upper state does not affect the probability of the transition at all. The longitudinal relaxation of the upper level accounts for the multiple returns of the particle to the lower state—each of which is accompanied by the emission of a spontaneous photon. In this case, the two-level system also comes back to the ground state in the middle of the Rabi cycle. But, these returns also do not bring any oscillations to the expectation value of the number of photons emitted per one atom. Gradual change of the quenching parameter results instead in the gradual transformation of Landau-Zener dependence to the smooth dependence given by the rate equation.

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