

Theory of fluctuational transitions between stable states of a nonlinear oscillator

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The path-integral method is used to calculate to logarithmic accuracy the general expression for the transition probability between stable states of a nonequilibrium system which is in interaction with a medium or is subjected to white noise. It is shown that in the first case the transition probability depends exponentially on $1/T$. As an example, transitions between the states of a nonlinear oscillator in the field of an external resonance force are considered. The calculation of the activation energy reduces to the solution of a variational problem. In the most interesting extreme cases explicit analytic expressions for the activation energy are derived, and its dependence on the parameters of the oscillator are analyzed. Characteristics of the absorption of an additional weak field are investigated, and also those of the absorption of the main field.

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Nonequilibrium systems often can have several states which are stable relative to small fluctuations. In this connection the problem arises of determining the probability of transitions between such states owing to relatively rare large fluctuations, and the distribution of systems over states.

One of the simplest systems of this kind is a nonlinear oscillator with an anharmonic term $\sim q^4$ in the potential, situated in a field of resonance force h . As is well known (cf. e.g., Refs. 1 and 2) when the amplitude of the force exceeds some critical value h_c [at which the nonlinear frequency shift $\Delta\omega(h_c)$ is of the order of the damping Γ] the oscillator can be in two stable vibrational states with different amplitudes. On the phase plane there are foci (or nodes) corresponding to these states; there is also a saddle point, through which the separatrix passes. If we can neglect the random action of the medium on the oscillator, then it is in one state or the other for an arbitrarily long time. Strictly speaking, however, the interaction with the medium, which causes friction, also leads to the appearance of a random force. This brings about transitions between the states.

The purpose of the present paper is to calculate the probability W of such transitions in the limiting case of a small random force, for which W is small in comparison with Γ . In this case after a time $\sim\Gamma^{-1}$ the system comes near one of the foci on the phase plane and fluctuates around it for a long time. Only rarely (after a time $\sim W^{-1}$) does the system experience a large enough fluctuation so that the trajectory crosses the separatrix. With overwhelming probability the crossing occurs near the saddle point. After this the system rapidly approaches a different focus, which means a transition to a new stable state. Obviously the probabilities of the transitions $1 \rightarrow 2$ and $2 \rightarrow 1$ are different, and their ratio determines the stationary distribution over the states.

The transition between stable states of a nonlinear oscillator is analogous in a certain sense to the transition of a diffusing particle through a potential barrier.

As is well known, for small W the thing of greatest interest in such problems is to calculate the coefficient in the exponent, and the pre-exponential factor often needs only to be found in order of magnitude. Here the transition probability between states of a nonequilibrium system will be calculated to logarithmic accuracy. For this purpose it is convenient to express W in the form of an integral over trajectories. The essential point is that the coefficient in the exponential is inversely proportional to the small parameter that characterizes the intensity of the random force. This allows us to reduce the problem of calculating W to the determination of the extremum of the coefficient in the exponential (see Sec. II). Actual calculations of $\ln W$ have been successfully done for ranges of values of the parameters which correspond to various extreme cases (see Secs. III and IV).

The system considered can serve as a model of certain physical objects, for example, of local or quasi-local vibrations near defects in crystals in strong resonance fields, of nonlinear oscillatory circuits in radiophysics, and of some nonlinear optical systems. In such problems it is of interest to determine the absorption coefficient of the external field. The resulting characteristic dependences on frequency and amplitude are examined in Sec. V. Features of the absorption of a weak additional field are also considered there. In particular, it is predicted that a very narrow absorption peak may appear with width $\sim W$, owing to induced transitions between foci.

The present problem is actually one example of a large class of problems of the behavior of nonequilibrium systems with some stable states. The method used, based on the application of continuous integration, is rather general and can be applied to various problems, if the transition probabilities are sufficiently small.

Since the calculation of W to logarithmic accuracy reduces to the determination of the mean time for reaching some boundary in phase space, for processes

of the Markov type the calculation of W can also be done in another way, based on the application of the Einstein-Fokker-Planck equation (see, e.g., Refs. 3-5). This method is convenient if the random process is one-dimensional, but for many-dimensional processes (in particular, for the two-dimensional processes considered here) its use is usually difficult. A special case of nonequilibrium systems comprises those in which the Einstein-Fokker-Planck equation can be solved exactly, for example, the Van der Pol oscillator in the field of an external random force.^{6,7} But in this last problem, for each particular set of parameters there is only one stable state.

After the present paper was prepared, we learned of interesting mathematical work of Wentzel and Freidlin,⁸ in which large fluctuations of dynamical systems caused by small random fluctuations are considered.¹⁾ In these papers a different method is used to find the probability of reaching a point of phase space remote from points that correspond to stable states. Equation (11) of Sec. I agrees with the results of Ref. 8 (but the problem of nonlinear oscillators was not treated there.⁸⁾ We note that the method of continuous integration can evidently be more convenient for actual research on more general problem, for example non-Markov cases, or those in which the number of dynamical variables is larger than the number of independent components of the random force.

1. FORCED VIBRATIONS OF A NONLINEAR OSCILLATOR INTERACTING WITH A MEDIUM

Let us consider a Duffing nonlinear oscillator which is in a field of periodic external force and interacts with a gas. We shall describe the medium with a set of continuous spectrum vibrations. The Hamiltonian of the system is

$$\begin{aligned} H &= H_0 + H_m + H_i; \\ H_0 &= \frac{1}{2}(p^2 + \omega_0^2 q^2) + \frac{1}{4}\gamma q^4 - qh \cos \omega \tau, \\ H_m &= \frac{1}{2} \sum_k (p_k^2 + \omega_k^2 q_k^2), \quad H_i = \sum_k \varepsilon_k q q_k. \end{aligned} \quad (1)$$

Here k numbers the vibrations of the continuous spectrum, and for simplicity it is assumed that the interaction with the medium is linear in the coordinate q of the individual nonlinear oscillator in question and in the coordinates q_k of the oscillators of the medium. The constants γ and ε_k are supposed sufficiently small that the renormalization of ω_0 owing to them is small. The frequency ω of the external force is assumed nearly equal to the proper frequency ω_0 .

For a statistical description of the motion of the oscillator under consideration we must eliminate the vibrations of the continuous spectrum. Solving the equations of motion of the q_k , we can express $q_k(\tau)$ in terms of $q(\tau')$ and the initial amplitudes and phases A_k and φ_k which are random quantities.⁹ After this the equation of motion for the oscillator considered takes the form

$$\begin{aligned} \frac{d^2 q}{d\tau^2} + \omega_0^2 q + \gamma q^3 &= h \cos \omega \tau + f(\tau) + L, \\ f(\tau) &= - \sum_k \varepsilon_k A_k \cos(\omega_k \tau + \varphi_k). \end{aligned} \quad (2)$$

Here $f(\tau)$ is the random force exerted by the medium on the oscillator, and L is an integral operator describing the reaction of the medium and allowing for retardation (cf. Ref. 9).

For small interaction with the medium the characteristic damping time of the oscillator, Γ^{-1} , is much larger than ω_0^{-1} and the characteristic period of the medium, ω_e^{-1} . For the analysis of the motion of the oscillator in the most interesting range of times $\tau \gg \tau_0$ [$\tau_0 = \max(\omega_0^{-1}, \omega_e^{-1})$] it is convenient to use a well known averaging method (cf., e.g., Ref. 2). We change from the rapidly oscillating functions $q(\tau), dq/d\tau$ to smoothed complex functions $u_1(\tau), u_2(\tau) = u_1^*(\tau)$:

$$q = u_1 e^{i\omega\tau} + u_2 e^{-i\omega\tau}, \quad dq/d\tau = i\omega(u_1 e^{i\omega\tau} - u_2 e^{-i\omega\tau}). \quad (3)$$

Dropping rapidly oscillating terms proportional to the small parameters γ, ε^2 , and using the asymptotic expression for L at large τ , in analogy with the procedure in Refs. 10 and 11 for the case $h=0$, we reduce the second-order equation to a complex equation of first order:

$$\begin{aligned} \frac{du_1}{d\tau} &= - \frac{3\gamma}{2i\omega} |u_1|^2 u_1 - (\Gamma \operatorname{sign} \tau + i\delta\omega) u_1 + \frac{h}{4i\omega} + \frac{1}{2i\omega} e^{-i\omega\tau} f(\tau), \\ \delta\omega &= \omega - \omega_0 - P. \end{aligned} \quad (4)$$

Explicit expressions for the damping Γ and the frequency shift $P(\Gamma, P \sim \varepsilon^2)$ are given in Refs. 10 and 11. In the derivation of Eq. (4) it has been assumed that

$$|\delta\omega|, \Gamma, |P|, \gamma |u_1|^2 / \omega \ll \omega_0, \omega_e, \quad |d\Gamma(\omega)/d\omega| \ll 1.$$

It is convenient to write Eq. (4) in dimensionless variables:

$$\begin{aligned} \dot{u} &= v + f, \quad v = v(u, u^*) = -u \operatorname{sign} t \\ &\quad + iu(uu^* \operatorname{sign} \gamma - \Omega) - i(\beta |\Omega|^2)^{1/2} \operatorname{sign} h; \\ u &= (3|\gamma|/2\omega\Gamma)^{1/2} u_1, \quad \dot{u} = du/dt, \quad t = \Gamma\tau, \quad \Omega = \delta\omega/\Gamma, \\ \beta &= \frac{3|\gamma|\hbar^2}{32\omega^3 |\delta\omega|^3}, \quad f(t) = -i \left(\frac{3|\gamma|}{8\omega^3 \Gamma^3} \right)^{1/2} \exp\left(-i \frac{\omega}{\Gamma} t\right) f\left(\frac{t}{\Gamma}\right). \end{aligned} \quad (5)$$

In Eq. (5) the complex random force $\bar{f} = f' + if''$ is δ -correlated (for $\tau \gg \tau_0$):

$$\begin{aligned} \langle \bar{f}'(t_1) \bar{f}'(t_2) \rangle &= \langle \bar{f}''(t_1) \bar{f}''(t_2) \rangle = 2\alpha \delta(t_1 - t_2), \\ \langle \bar{f}'(t_1) \bar{f}''(t_2) \rangle &= 0, \quad \alpha = 3|\gamma|\hbar^2 / 8\omega^3 \Gamma^3. \end{aligned} \quad (6)$$

Equation (5), with the random force f having the statistical properties (6) is valid also for a more general form of the weak-interaction Hamiltonian H_i containing terms nonlinear in the q_k , and also when an interaction between the vibrations of the medium has to be taken into account. The only change is in the explicit expressions for Γ and P .¹⁰

By means of the change of variables (3) one can also reduce to Eq. (5) the problem of the forced vibrations of a nonlinear Brownian oscillator in the field of an arbitrary random δ -correlated force f_1 (not necessarily due to thermal fluctuations):

$$\begin{aligned} \frac{d^2 q}{d\tau^2} + 2\Gamma \frac{dq}{d\tau} + \omega_0^2 q + \gamma q^3 &= h \cos \omega \tau + f_1(\tau), \\ \langle f_1(\tau) f_1(\tau') \rangle &= 2B\delta(\tau - \tau'). \end{aligned} \quad (7)$$

The parameter $\alpha = 3|\gamma|B/16\omega^3\Gamma^2$ in Eq. (6) is now determined by the noise characteristic B . Actually Eq. (5) is the Langevin equation for a nonlinear damped oscillator.

In the absence of the random force the complex nonlinear equation of motion (5) has stationary solutions ($\dot{u} = 0$) whose amplitude is determined by the equation

$$\varphi(|u|^2) = 0, \quad \varphi(x) = x(x \operatorname{sign} \gamma - \Omega)^2 + x - \beta|\Omega|^3. \quad (8)$$

For values of the parameters β and Ω that lie within the region in Fig. 1 bounded by the solid curves, Eq. (8) has three solutions, and if the point β, Ω is outside this region it has one solution. Two of the three solutions are stable (with largest and smallest amplitude values $|u|$), and the third solution corresponds to a saddle point. The separatrix goes through this point $u', u'' (u = u' + u'')$ on the phase surface.

The random force in Eq. (5) makes the motion stochastic. For a small random force the system spends nearly all of its time near one focus or another, fluctuating around it with an amplitude $\langle \delta u^2 \rangle^{1/2} \sim \alpha^{1/2}$ which is much smaller than the distance between the focus and the separatrix. There is a very small probability of the occurrence of large fluctuations which lead to crossing of the separatrix and transition of the system into a different stable state.

2. GENERAL EXPRESSION FOR THE TRANSITION PROBABILITY

Let us consider the probability density $w(t_a, u_a; t_b, u_b)$ for the realization of trajectories that pass at time t_a through a point u_a on the phase surface within the region of attraction to a focus f , and at time t_b through a point u_b near the separatrix. It is convenient to write this quantity as a continuous integral:

$$w(t_a, u_a; t_b, u_b) = \int Df(t) P[f(t)] \delta(u(t_a) - u_a) \times \delta(u(t_b) - u_b) \left\{ \int Df(t) P[f(t)] \right\}^{-1}. \quad (9)$$

Here $Df(t) \equiv D\tilde{f}'(t) D\tilde{f}''(t)$. The functional P determines the probability distribution of the random function $f(t)$. As is well known (cf. Ref. 12), for a random function of the type of white noise with the correlation (6) this

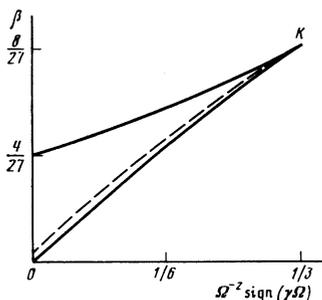


FIG. 1. Region of existence of two stable states of a nonlinear oscillator.

functional is

$$P[f(t)] = \exp \left[-\frac{1}{4\alpha} \int |f(t)|^2 dt \right]. \quad (10)$$

We are interested in calculating the probability (9) in the region of times $(W/\Gamma)^{-1} \gg t_b - t_a \gg 1$ ($\tau_b - \tau_a \gg \Gamma^{-1}$), where W is the probability per unit time of transition from f to a different focus f' . Moreover, during a time $t - t_a \sim 1$ the system arrives near the focus f and forgets the initial state u_a . The displacement to the point u_b also comes about quickly, during a time ~ 1 , from some point near the focus f (though it is improbable).

Subject to conditions to be indicated later on, in the range $t_b - t_a \gg 1$ the transition probability (9) is exponentially small. It has the small parameter α in the denominator of the coefficient in the exponent. Making the calculation to logarithmic accuracy, we shall determine only the exponent and not concern ourselves with the pre-exponential factor, which depends only weakly on α . In Eq. (9), following Ref. 12, it is convenient to change from integration over the random force, $D\tilde{f}(t)$, to integration over the trajectories of the nonlinear oscillator, $Du(t)$. It can be seen from Eqs. (5) that the Jacobian for the transformation does not depend on α and affects only the pre-exponential factor, which is of no interest to us. To the accuracy envisaged we also can deal in the integral over trajectories with just the main exponential factor, corresponding to the extremal trajectory. Then, in view of Eqs. (10) and (5), the probability (9) is given by

$$w(t_a, u_a; t_b, u_b) = \text{const} \cdot \max \exp(-S/4\alpha), \quad S = \int_{t_a}^{t_b} \mathcal{L}(u, \dot{u}) dt, \quad \mathcal{L}(u, \dot{u}) = |\dot{u} - v(u, u^*)|^2, \quad u(t_a) = u_a, \quad u(t_b) = u_b. \quad (11)$$

Here t_c is the time when the large fluctuation starts, which shifts the system to u_b ; u_c is a point in a small neighborhood of the focus ($|u_c - u_f| \lesssim \alpha$); and the maximum is taken with respect to possible trajectories and to $t_b - t_c$. Actually there should be an integration over u_c , with an appropriate distribution function, in Eq. (11), but this would affect only the pre-exponential factor.

The quantity $\mathcal{L}(u, \dot{u})$ in Eq. (11) can be regarded as the Lagrangian of a particle, and S , as its action. Since \mathcal{L} does not depend on the time explicitly, we have $\partial S / \partial t = -E$, where E is the energy of the particle (cf. Ref. 1), and the condition that S in Eq. (11) be extremal with respect to $t_b - t_c$ reduces to the equation

$$E = |\dot{u}|^2 - |v(u, u^*)|^2 = 0. \quad (12)$$

The probability W for transitions between states in neighborhoods of foci f and f' is given to within a factor $\sim \frac{1}{2}$ by the integral of the expression (11) over points u_b in the neighborhood of the separatrix; in our approximation we are to find the extremum with respect to u_b in this neighborhood. Since $\partial S / \partial u_b = \partial \mathcal{L} / \partial u$, we have

$$\delta S = (\dot{u}^* - v^*) \delta u_b + (\dot{u} - v) \delta u_b^*.$$

For a shift of the point u_b along the separatrix $\delta u_b / v = \delta u_b^* / v^*$; therefore, in view of Eq. (12), S

$= -|\dot{u} - v|^2 v^{-1} \delta u_b$. If the point u_s shifts in the direction of the saddle point, then $v^{-1} \delta u_b > 0$, and consequently among the trajectories ending on the separatrix the extremal ones will be those that go through the saddle point u_s . At this point $v = 0$, and according to Eq. (12) a particle near it moves slowly ($t_b - t_c \rightarrow \infty$ for $u_b \rightarrow u_s$). In the neighborhood of the saddle point small variations from the extremal trajectory become important, having a probability $\sim \frac{1}{2}$ of taking the system across the separatrix in a time $\Delta t \sim 1$. Therefore, to accuracy up to the preexponential factor we can write W in the form

$$W = \text{const} \cdot \exp(-Q/\alpha), \quad Q = \frac{1}{2} \min \int |\dot{u} - v|^2 dt, \quad (13)$$

$$u(0) \approx u_1, \quad u(t) \approx u_2.$$

The condition for applicability of Eq. (13) is the inequality

$$Q \gg \alpha. \quad (14)$$

Equation (11) can also be derived from the results of Ref. 8. We note that the method used here, of changing a continuous integral over random forces into one over random variables, can also be used to consider large fluctuations in more complicated systems.

For the δ -correlated random force considered here the characteristic parameter for its intensity appears in the expression (13) for the transition probability only as the factor $1/\alpha$ in the exponent. In the case of a nonlinear oscillator interacting with a medium, we see from Eq. (6) that $\alpha \sim T$, i.e., the transition probability depends on $1/T$ (the activation energy being $Q kT/\alpha$). This result holds also for other subsystems interacting with a medium and described by a kinetic equation.

3. TRANSITION PROBABILITIES WITH A RELATIVELY LARGE FREQUENCY MISMATCH

In the expression (13) for the transition probability the quantity Q depends only on the dynamical characteristics of the subsystem. In the present case of a nonlinear oscillator, according to Eqs. (5) and (13) Q is a function of two dimensionless parameters, β and Ω . For any specific values of β and Ω , Q can be calculated by numerical solution of the Euler equation for the variational problem (13), (12). In limiting cases explicit analytic expressions can be obtained for $Q(\beta, \Omega)$. This can be done if $|\Omega| \gg 1$ or if the values of β and Ω are close to the critical values and lie near the point K in Fig. 1. These respective limiting cases are considered in this section and the following one.

Since two stable states of the oscillator exist only for $\gamma \Omega > 0$, and Q does not depend on the signs of γ and h , we shall from now on suppose that $\gamma > 0, \Omega > 0, h > 0$. In the case $\Omega \gg 1$ it is convenient to change from u, u^* to real variables x, y and from t to a new time t' :

$$x = \Omega^{-1/2} \text{Re } u, \quad y = \Omega^{-1/2} \text{Im } u, \quad (15)$$

$$t' = \Omega t, \quad \dot{x} = dx/dt'$$

(from now on differentiation with respect to t' will be denoted with a dot). In these variables the expressions (13) for Q and (12) for E take the form

$$Q = \frac{1}{4\epsilon^2} \min \int_0^{t'} dt' [(x + g_v + \epsilon x)^2 + (y - g_x + \epsilon y)^2], \quad (16)$$

$$E = \epsilon^{-2} [x^2 + y^2 - (g_v + \epsilon x)^2 - (g_x - \epsilon y)^2] = 0,$$

$$g(x, y) = \frac{1}{2} (x^2 + y^2 - 1) - x\sqrt{\beta}, \quad \epsilon = \Omega^{-1}, \quad g_x = \partial g / \partial x,$$

and the extremal trajectories are described by the following Euler equations:

$$\ddot{x} + (g_{xx} + g_{vv})\dot{y} - \frac{1}{2} \frac{\partial}{\partial x} (g_x^2 + g_v^2) - \epsilon^2 x = 0, \quad (17)$$

$$\ddot{y} - (g_{xx} + g_{vv})\dot{x} - \frac{1}{2} \frac{\partial}{\partial y} (g_x^2 + g_v^2) - \epsilon \sqrt{\beta} - \epsilon^2 y = 0.$$

Equations (17) contain linear and quadratic terms in the small parameter ϵ . In the Appendix a self-similar solution of these equations is found by means of asymptotic perturbation theory (in the range of times $t' \sim \epsilon^{-1}$) with proper allowance for the "nonstandard" situation—degeneracy of the "fast" motion. The first-order (in ϵ) corrections are determined from a secular equation which is found by considering second-order terms. The result for the quantity Q , which determines, according to Eq. (13), the transition probability, is given by the expression (A.13) in the form of a quadrature. It can be represented in analytic form for small β . According to Eqs. (A.13), (A.15) the values Q_1 for the transition from focus f_1 (which corresponds to the smaller amplitude of the oscillations) and Q_2 for that from focus f_2 are given by the formulas

$$\epsilon Q_1 = \frac{1}{2} - \xi \beta^{3/4}, \quad \epsilon Q_2 = 2\sqrt{\beta}, \quad \epsilon^2 \ll \beta \ll 1. \quad (18)$$

Here the constant ξ can be expressed as a quadrature of an elliptic integral, $\xi \approx 0.98$. If the parameter β is not small (but, as before, $\epsilon = \Omega^{-1} \ll 1$), Q_1 and Q_2 can be calculated numerically from Eqs. (A.11) and (A.13). The results of such calculations are shown in Fig. 2 [the curves (18) are shown with dashed lines]. From Eqs. (13), (A.13), and (18) and from Fig. 2 we see the dependence of the factor Q/α in the exponent on the parameters of the oscillator. Q/α depends on the external field only through the parameter $\beta \sim h^2/|\delta\omega|^3$; Q_1 decreases monotonically with increasing field, and Q_2 increases. The frequency mismatch $\delta\omega$ occurs in both β and Ω ; for fixed β we have from Eq. (A.13) $Q \sim |\delta\omega|$. For $\Gamma \ll \delta\omega$ the damping parameter Γ drops out of the expression for Q/α altogether. In the present case $\epsilon \ll 1$ the quantity q/α is proportional not only to $\alpha^{-1} \gg 1$, but also to $\epsilon^{-1} \gg 1$.

The ratio of the probabilities w_1 and w_2 for finding the

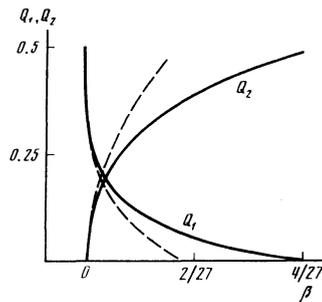


FIG. 2. Dependence of Q_1 and Q_2 on β for $\Omega \gg 1$.

system at the foci f_1 and f_2 is given by [Eq. (13)]:

$$w_1/w_2 = W_{21}/W_{12} = \text{const} \cdot \exp[(Q_1 - Q_2)/\alpha]. \quad (19)$$

To logarithmic accuracy, as Fig. 2 shows, for $\beta_0 - \beta \gg \alpha \varepsilon$ ($\beta_0 \approx 0.013$) we have $w_2 \ll w_1 \approx 1$, and for $\beta - \beta_0 \gg \alpha \varepsilon$ we have $w_1 \ll w_2 \approx 1$. In the region $|\beta - \beta_0| \sim \alpha \varepsilon$ the two probabilities are of the same order of magnitude.

Special consideration has been given to the case $\beta \approx \varepsilon^2 \ll 1$ (while $\beta - \varepsilon^2 + \varepsilon^3/4 \geq 0$). It can be shown that in this case

$$\varepsilon Q_1 \approx 1/2, \quad \varepsilon Q_2 = \xi_1 (\chi/\varepsilon) \beta^{1/2} \chi^3, \quad \xi_1 (\chi/\varepsilon) \sim 1,$$

$\chi^2 = (\beta - \varepsilon^2)/\varepsilon^2 + \beta/4$, $\chi \ll 1$; for $\chi \ll \varepsilon \approx \beta^{1/2} \ll 1$, $\xi_1 \approx 2/3$. It can be seen that εQ_2 depends on β and on the ratio of the two small parameters β and χ (for $\chi \rightarrow 0$ the second focus merges with the saddle point and disappears).

4. TRANSITION PROBABILITIES NEAR THE CRITICAL POINT

Analysis of the Euler equations (17) and calculation of Q can also be simplified for values of the parameters $\beta \approx \beta_c = 8/27$, $\Omega \approx \Omega_c = 3^{1/2}$, which are near the point K in Fig. 1. If $\beta = \beta_c$ and $\Omega = \Omega_c$, the two nodes and the saddle point all coincide, and according to Eqs. (5) and (15) they are at $x_c = -1/6^{1/2}$, $y_c = -1/2^{1/2}$. For $\beta \approx \beta_c$ and $\Omega \approx \Omega_c$ the distance between the nodes is small and it is convenient to introduce new variables:

$$x_1 = x - x_c, \quad y_1 = y - y_c, \quad v = 1 - \Omega_c/\Omega, \quad (20)$$

$$\Delta = 9\beta_c (1 - \sqrt{\beta/\beta_c}) - 2v.$$

In these variables the expression (16) for Q takes the form

$$Q = \frac{3}{4} \min \int_0^1 [(x_1 - \zeta)^2 + (y_1 - \eta)^2] dt',$$

$$\zeta = -\frac{2}{3} (3^{1/2} x_1 + y_1) + \frac{1}{3\sqrt{2}} (3^{1/2} x_1 + y_1)^2$$

$$+ \frac{4\sqrt{2}}{3} y_1^2 - y_1^3 - x_1^2 y_1 - \frac{v}{3\sqrt{2}} + \frac{v x_1}{\sqrt{3}}$$

$$\eta = -\frac{1}{\sqrt{6}} (3^{1/2} x_1 + y_1)^2 + x_1^3 + x_1 y_1^2 + \frac{v y_1}{\sqrt{3}} + \frac{\Delta}{2\sqrt{6}}. \quad (21)$$

Since for $\beta \approx \beta_c$, $\Omega \approx \Omega_c$ the distance between the nodes is $\sim \nu^{1/2} \ll 1$, on the extremal trajectories x_1, y_1 are also $\sim \nu^{1/2}$. Analyzing the Euler equations for the problem (21) for small ν and using the law of the conservation of energy (16), we can show that the characteristic time is proportional to ν . Owing to this, in the Euler equations

$$\ddot{x}_1 + (\eta_{x_1} - \zeta_{x_1}) \dot{y}_1 - \frac{1}{2} \frac{\partial}{\partial x_1} (\zeta^2 + \eta^2) = 0,$$

$$\ddot{y}_1 - (\eta_{y_1} - \zeta_{y_1}) \dot{x}_1 - \frac{1}{2} \frac{\partial}{\partial y_1} (\zeta^2 + \eta^2) = 0, \quad (22)$$

the terms with \ddot{x}_1 or \ddot{y}_1 are of order $\nu^{5/2}$, and those with \dot{x}_1 or \dot{y}_1 are of order $\nu^{3/2}$. Therefore in the formula for ζ the terms $\sim \nu^{1/2}$ and $\sim \nu$ must be set equal to zero, so that we can express $x_1 = -y_1/3^{1/2} + \dots$ in terms of y_1 with accuracy to terms $\sim \nu$. Substituting this expression for x_1 in Eq. (22) and considering that $\zeta \sim \eta \sim \nu^{3/2}$, $\zeta_{y_1} \sim 1$, $\eta_{x_1} \sim \eta_{y_2} \sim \nu$, we find that $\dot{x}_1 = \zeta + 0(\nu^2)$. Then from the law of conservation of energy

$$\dot{x}_1^2 + \dot{y}_1^2 - \zeta^2 - \eta^2 = 0$$

we get for $x_1 \approx -y_1/3^{1/2}$

$$\dot{y}_1 = \pm \tilde{\eta}(y_1) + O(\nu^2),$$

where

$$\tilde{\eta}(y_1) = \eta\left(x_1 = -\frac{y_1}{\sqrt{3}}, y_1\right) = \frac{1}{\sqrt{3}} y_1 \left(v - \frac{4}{3} y_1^2\right) + \frac{\Delta}{2\sqrt{6}}$$

(in the region of existence of two nodes near the point K , $\Delta \lesssim \nu^{3/2}$). The expression $\dot{y}_1 = \pm \tilde{\eta}(y_1)$ can also be derived by solving Eq. (22) with perturbation theory including terms $\sim \nu^{5/2}$ and not using the conservation of energy.

The motion from the node to the saddle point is described by the solution with $\dot{y}_1 = -\tilde{\eta}$. Substituting it in Eq. (21) and considering that $\dot{x}_1 \approx \zeta$, we get

$$Q = -3 \int_{\nu'}^{\nu''} \tilde{\eta}(y_1) dy_1 = \frac{\sqrt{3}}{4} v^2 F(\bar{\Delta}), \quad \bar{\Delta} = \frac{3\Delta}{2\sqrt{2}v^{3/2}} \quad (23)$$

$$F(\bar{\Delta}) = z_f(z_f + \bar{\Delta}) - z_s(z_s + \bar{\Delta}), \quad z_{f,s}^3 - \nu^{3/2}/z_{f,s} - \nu^{1/2} \bar{\Delta} = 0.$$

Here z_s and z_f denote the middle root and one of the extreme roots of the given cubic equation and determine the amplitudes of the oscillator at the saddle point and at the nodes. The parameter $\bar{\Delta}$ lies in the interval $-1 \leq \bar{\Delta} \leq 1$. The function $F_1(\bar{\Delta})$ for the transition from the node f_1 (with the smallest amplitude) increases monotonically with $\bar{\Delta}$ from the value $F_1(-1) = 0$ to $F_1(1) = 9/4 [F_1(0) = 3/4]$ and $F_2(\bar{\Delta}) = F_1(-\bar{\Delta})$.

It can be seen from Eq. (23) that as the critical point is approached the activation energy falls off as $\nu^2 \sim (\Omega - \Omega_c)^2$. For $\bar{\Delta} > 0$ we have $Q_1 > Q_2$; that is, the state is more probably near the node f_1 , while for $\bar{\Delta} < 0$ it is more probably near f_2 . Therefore the line $\beta = \beta_0$ on which the two states are equally probable is given for $\Omega \approx \Omega_c$ by the condition $\Delta = 0$, and by (20)

$$\beta_0 = \beta_c (1 - \nu^{1/2}).$$

Using this result for the section of the curve $\beta_0(\Omega^{-2})$ with $\Omega \approx \Omega_c$ and the previously obtained value of β_0 for $\Omega^{-1} \rightarrow 0$, and also the monotonic nature of the dependence of β_0 on Ω^{-2} , which is intuitively obvious, we can get the interpolated curve of $\beta_0(\Omega^{-2})$ shown by the dashed curve in Fig. 1.

5. FEATURES OF THE ABSORPTION COEFFICIENTS OF A STRONG FIELD AND AN ADDITIONAL WEAK FIELD

The dynamical characteristics of a nonlinear oscillator must have specific features for relations between the field h and the frequency mismatch $\delta\omega$ such that $\beta \approx \beta_0$ and the probabilities w_1 and w_2 of the stable states are comparable. In this region there are changes both of the absorption coefficient of the strong resonance field h and of the absorption coefficient of an additional weak field. In our analysis of these effects we shall consider the case in which the time of observation is much larger than the transition times W_{12}^{-1} and W_{21}^{-1} and that there is no hysteresis.^{1,2}

The power I drawn from the strong field is given by

$$I = m \left\langle \frac{dq}{dt} h \cos \omega \tau \right\rangle = -m \omega h \sum_{i=1}^2 w_i \text{Im } u_{ij},$$

where m is the generalized mass of the oscillator. The

imaginary part of u_{f_i} at the focus f_i ($i=1, 2$) can be expressed in terms of $|u_{f_i}|^2$ by equating $v(u, u^*)$ to zero in Eq. (5). After this we get for the quantity $\mu = 2I/h^2$, which determines the absorption coefficient, the value

$$\mu = \frac{m\Gamma^2}{2\beta|\delta\omega|^2} (\omega_1|u_{f_1}|^2 + \omega_2|u_{f_2}|^2), \quad (24)$$

where $|u_{f_i}|^2$ are roots of the cubic equation (8).

Equation (24) enables us to follow the dependence of μ on $\delta\omega$ and h . In the region $\beta \approx \beta_0$ with a relatively small change of the parameters the quantity in parentheses in Eq. (24) changes sharply [exponentially rapidly in accordance with Eq. (19)] from $|u_{f_1}|^2$ to $|u_{f_2}|^2$ (or vice versa). Furthermore the curve of μ as a function of h must show a somewhat smeared out kink, and on both sides of this $\mu(h)$ falls off monotonically, if $\beta_0 \geq \Omega^{-2}$.

Besides the absorption of the strong field at the frequency ω it can be of interest to study the absorption of an additional weak field h' at a different frequency ω' . To determine the corresponding quantity $\mu' = 2I'/h'^2$ in the resonance region $|\omega' - \omega| \sim |\omega - \omega_0|$ we can insert a term $h'\cos\omega't$ in Eq. (2) and solve Eq. (4) with the term in h' included, linearizing it with $\delta u_i \sim h'$. Then

$$\mu' = \sum_{i=1}^2 w_i \mu'_i, \quad \mu'_i = \frac{m}{2\Gamma} \frac{1 + (\Omega - \Omega' - 2|u_{f_i}|^2) - |u_{f_i}|^4}{(\lambda_i^2 - \Omega'^2) + 4\Omega'^2}, \quad (25)$$

$$\Omega' = (\omega' - \omega)/\Gamma, \quad \lambda_i^2 = \Omega^2 - 4\Omega|u_{f_i}|^2 + 3|u_{f_i}|^4 + 1.$$

According to Eq. (25), the spectrum of $\mu'(\Omega')$ consists of two partial spectra μ'_i , which are comparable in intensity for $\beta \approx \beta_0$. The structure of the spectra $\mu'_i(\Omega')$ is determined by the values of $|u_{f_i}|^2$. For $|u_{f_i}|^2 > 1$ the spectrum $\mu'_i(\Omega')$ has two maxima separated by a minimum, at which $\mu'_i < 0$, i.e., the weak field is strengthened at the expense of the strong field (but the absorption integrated over the frequency Ω' is positive and equal to $\pi m/2\Gamma$). If $|u_{f_i}|^2 \gg 1$ (then $\lambda_i^2 \gg 1$) the widths of the peaks are smaller than the distance between them by about factors λ_i .

In the region $|\omega' - \omega| \sim W_{21} + W_{12}$, besides the absorption (25) a term $\bar{\mu}'$ becomes important; this term is due to transitions between the foci which are induced by the weak field. It can be shown that

$$\bar{\mu}' = \frac{m}{\Gamma} \text{Im} \left[C(u_{f_1} - u_{f_2}) \frac{w_1 W_{12} + w_2 W_{21}}{W_{12} + W_{21} + i\Omega\Gamma} \right], \quad |C| \sim 1. \quad (26)$$

The absorption $\bar{\mu}'(\omega')$ has a very sharp maximum (of width much smaller than the characteristic width Γ) in the region $\Omega' = 0$ ($|\omega' - \omega| \ll \Gamma$), and is exponentially small outside a narrow region $\beta \approx 0$. If there is a q^3 term in the Hamiltonian of the oscillator, the mean dipole moments of the stable states are different and an analogous peak of relaxation type appears at low frequencies $\omega' \approx W_{12} + W_{21}$. These questions will be considered in more detail elsewhere.

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APPENDIX

It is convenient to construct the asymptotic solution of the equations (17) by starting from functions X, Y

which satisfy the equations

$$\dot{X} = -g_x, \quad \dot{Y} = g_x, \quad g = g(X, Y), \quad (A.1)$$

since, in zeroth approximation in ε , $x = X$ and $y = Y$ is a solution of Eq. (17) and makes the large terms in Eq. (16) cancel. The equations (A.1) have the form of Hamilton's equations for a particle executing a one-dimensional finite motion with the Hamiltonian g . This motion is periodic with frequency $\omega(g) \sim 1$ and is described by functions periodic in the phase ψ :

$$X = X(g, \psi) = X(g, \psi + 2\pi), \quad Y = Y(g, \psi) = Y(g, \psi + 2\pi), \\ g = \text{const}, \quad \dot{\psi} = \omega(g).$$

In what follows we look for a solution of Eq. (17) (for $\varepsilon \neq 0$) in the form $x = X(g, \psi)$, $y = Y(g, \psi)$, supposing, however, that g and $\psi - \omega(g)$ depend on the time. Actually this amounts to a well known (cf., e.g., Refs. 2, 13) transition from x, y to variables g, ψ , one of which changes slowly with time ($\dot{g} \sim \varepsilon$). We introduce the auxiliary functions A and B by means of the equations

$$\dot{x} = \dot{X} = \omega(g) X_\psi - \varepsilon X + \varepsilon A(g, \psi), \quad \dot{\omega}(g) X_\psi = -g_x, \\ \dot{y} = \dot{Y} = \omega(g) Y_\psi - \varepsilon Y + \varepsilon B(g, \psi), \quad \dot{\omega}(g) Y_\psi = g_x. \quad (A.2)$$

The functions A and B do not depend on \dot{g} nor on $\dot{\psi} - \omega(g)$ because the extremal trajectories in the phase space x, y are not self-intersecting.

We shall derive equations for the functions A and B whose solution in zeroth order in ε contains an arbitrary function. It will be determined in such a way that in the higher orders of perturbation theory A and B will contain no terms secular in ψ .

In Eq. (A.2) let us express \dot{X} and \dot{Y} in terms of \dot{g} , $\dot{\psi}$ and use the identity

$$X_\psi Y_\psi - X_\psi Y_\psi = \omega^{-1}(g) (g_x X_\psi + g_y Y_\psi) = \omega^{-1}(g). \quad (A.3)$$

Then

$$\dot{g} = \varepsilon \omega [(A - X) Y_\psi - (B - Y) X_\psi], \quad \dot{\psi} = \omega + \varepsilon \omega [(B - Y) X_\psi - (A - X) Y_\psi] \quad (A.4)$$

[here and in the following formulas $\omega \equiv \omega(g)$].

Differentiating the expressions (A.2) with respect to time by using Eqs. (A.4) and substituting in Eq. (17), we get equations for the functions A and B :

$$\omega A_\psi - 2XYA + (3X^2 + Y^2 - 1)B = -A_\psi \dot{g} - A_\psi (\dot{\psi} - \omega) + \varepsilon A, \\ \omega B_\psi - (X^2 + 3Y^2 - 1)A + 2XYB = -B_\psi \dot{g} - B_\psi (\dot{\psi} - \omega) + \varepsilon B. \quad (A.5)$$

Substituting Eq. (A.2) in the equation (16) for E , we can find an additional relation between A and B , which is one of the integrals of the equations of motion (A.5) with a constant corresponding to $E = 0$:

$$2g_y A - 2g_x B = \varepsilon [(A - X)^2 + (B - Y)^2 - X^2 - Y^2]. \quad (A.6)$$

In first order in ε the right members of Eqs. (A.5) and (A.6) are equal to zero. Using Eq. (1.6) and one of the equations (A.5) and taking into account the relations (A.2) between X_ψ , Y_ψ and g_x, g_y , we can find a general integral of the corresponding linear partial differential equation which contains one arbitrary function $F(g)$:

$$A^0 = F(g) Y_\psi, \quad B^0 = -F(g) X_\psi, \\ A = A^0 + \varepsilon A', \quad B = B^0 + \varepsilon B'. \quad (A.7)$$

The function $F(g)$ can be determined from the condition that the second order terms in εA and εB not contain terms secular in ψ , i.e., that these terms remain small all the way up to times $t' \sim \varepsilon^{-1} \gg \omega^{-1}(g) \sim 1$. In the second approximation we must replace A and B in the right members of Eqs. (A.5) and (A.6) with A^0 and B^0 from Eq. (A.7). Expressing B' in terms of A' by means of Eq. (A.6) and substituting in the first of the equations (A.5), we get a linear equation for A' :

$$\begin{aligned} \omega A_\psi' + (g_{xx} g_\psi g_x^{-1} - 2XY) A' &= Y_\psi P, \\ P &= -Y_\psi^{-1} [\varepsilon^{-1} A_\psi^0 \dot{g} + \varepsilon^{-1} A_\psi^0 (\psi - \omega) - A] + g_{xx} (2g_x Y_\psi)^{-1} R, \\ R &= (A^0)^2 + (B^0)^2 - 2A^0 X - 2B^0 Y = F^2(g) (X_\psi^2 + Y_\psi^2) - 2F(g) (XY_\psi - YX_\psi). \end{aligned} \quad (\text{A.8})$$

Here \dot{g} and ψ are determined by Eqs. (A.4) and (A.7).

Equation (A.8) can be solved by the method of variation of the arbitrary constant $D = D(g, \psi)$:

$$A' = D(g, \psi) Y_\psi, \quad \omega \frac{\partial D}{\partial \psi} = P. \quad (\text{A.9})$$

The periodicity of X and Y in ψ makes all the terms in P periodic. To assure that A' contains no terms secular in ψ , according to Eq. (A.9) we must have $P = 0$, where the bar indicates the average over ψ from 0 to 2π . The expression (A.8) for P can be put, after some long manipulations using Eqs. (A.4), (A.7), and (A.3), into the form

$$P = \frac{1}{2} \frac{\partial \omega}{\partial g} R - \frac{1}{2} \omega \frac{\partial R}{\partial g} + \frac{\partial Z(g, \psi)}{\partial \psi}. \quad (\text{A.10})$$

The explicit form of the function Z is not needed, since it drops out in the averaging over ψ . Then the condition $P = 0$ reduces to a simple equation for R , and the solution is $R = c\omega(g)$.

Near a focus $g_x, g_y \sim |g - g_f|^{1/2}$, where g_f is the value of g at the focus, and $\omega(g_f) \neq 0$. Therefore in order for B' to remain finite near the focus, according to Eq. (A.6) [where the right-hand member is equal to $\varepsilon R + O(\varepsilon^2)$] the constant c must be equal to zero.

From the formula (A.8) and R it follows that

$$\bar{R}(g) = F^2 m_1 - 2F m_2, \quad m_1(g) = \bar{X}_\psi^2 + \bar{Y}_\psi^2, \quad m_2(g) = \overline{XY_\psi - YX_\psi}. \quad (\text{A.11})$$

It can be seen from this that the condition $R = 0$ leads to two possible values of F and thus to two values of g in Eq. (A.4):

$$F_1 = 0, \quad F_2 = 2m_2/m_1, \quad \bar{g}_{1,2} = \mp \varepsilon \omega m_2. \quad (\text{A.12})$$

According to Eqs. (A.7) and (A.2) the solution $F_1 = 0$ describes motion in the absence of the random force from the saddle point to the focus: $\text{sign } \dot{g}_1 = -\text{sign}(g_1 - g_f)$, since according to Eqs. (A.3) and (A.11)

$$m_2(g) = 2 \int_{g_f}^g \omega^{-1}(g) dg.$$

It is obvious that the second solution F_2 satisfies the boundary conditions (13); this solution corresponds to motion from the focus to the saddle point. Furthermore according to Eqs. (16), (A.2), (A.7), and (A.12)

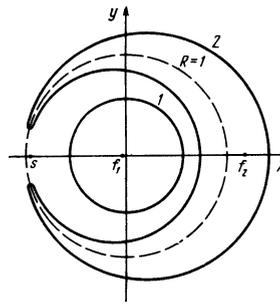


FIG. 3.

the quantity Q in Eq. (13) is given by

$$Q = \frac{1}{4} \int_0^{t'} F^2 (X_\psi^2 + Y_\psi^2) dt' \approx \frac{1}{\varepsilon} \int_{g_f}^{g_s} \frac{m_2}{\omega m_1} dg. \quad (\text{A.13})$$

Here g_s is the value of g at the saddle point, and in changing to the integration over g Eq. (A.12) was used.

Using the definition (A.11) of m_1 and m_2 and the formulas (A.2) for ωX_ψ and ωY_ψ , we can write the quantity $m_2/\omega m_1$ in the form

$$\begin{aligned} m_2/\omega m_1 &= \oint (X dY - Y dX) / \oint (-g_y dX + g_x dY) \\ &= 2 \iint dX dY / \iint \Delta g dX dY; \quad \Delta g = 4R^2 - 2. \end{aligned} \quad (\text{A.14})$$

Here $R^2 = X^2 + Y^2$, and the double integrals are taken over the region bounded by a trajectory $g(X, Y) = \text{const}$.

Explicit expressions for $m_2/\omega m_1$ can be obtained for small β . For $\varepsilon^2 \ll \beta \ll 1$, using the expression (16) for g , we have from (A.1): $g_{f_1} \approx \frac{1}{4}$, $g_s \approx -g_{f_2} \approx \beta^{1/2}$ (the positions of the foci and the saddle point are shown in Fig. 3). Near the focus f_1 the trajectories $g(X, Y) = \text{const}$ for $\beta \ll 1$ close to a circle of radius given by $R^2 \approx 1 - 2g^{1/2}$ (see curve 1 in Fig. 3), and near f_2 they are horseshoe-shaped, going around the circle $R = 1$ (see curve 2 in Fig. 3). For trajectories 1 and 2 we get from Eqs. (A.14) and (16), respectively, the formulas

$$\begin{aligned} \left(\frac{m_2}{\omega m_1} \right)_1 &= -\pi \left[\int_0^{2\pi} d\varphi (g + \beta^{1/2} \cos \varphi)^{1/2} \right]^{-1}, \quad \left(\frac{m_2}{\omega m_1} \right)_2 \\ &= 1, \quad \beta \ll 1. \end{aligned} \quad (\text{A.15})$$

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Nonresonant transitions and ionization of atoms in slow collisions in a laser field

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We consider inelastic atomic collisions that take place in the field of intense electromagnetic radiation and are accompanied by absorption of nonresonant photons. The cross sections are calculated for the excitation $[H(1S) + H(1S) + \omega_1 \rightarrow H(1S) + H(2S)]$ and charge exchange $[H(1S) + H(1S) + \omega_2 \rightarrow H^+ + H^-]$ of hydrogen atoms in close collisions. The transition cross sections turn out to depend substantially on the form of the potential curves of the quasimolecule H_2 . We calculate also the two-photon ionization of hydrogen atoms irradiated by an intense laser pulse in close collisions. The cross section depends little on the form of the potential curves of the quasimolecule, and an estimate of its value shows that collision ionization makes a contribution comparable with that of direct ionization even at medium densities. In the case of remote collisions, an analytic expression is obtained for the cross section of nonresonant transitions following adiabatic turning on the field, with account taken of the Stark shift of the atomic levels.

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1. INTRODUCTION

Much attention has been paid in recent years to the study of the influence of intense electromagnetic radiation on the dynamics of atomic collisions. Interest in this group of problems is due to the possibility of using laser radiation to stimulate various processes that occur in atomic collisions. Most theoretical and experimental studies were devoted to the so-called optical and radiative collisions (see the review by Yakovlenko¹), which occur at large interatomic distances, and the frequency of the absorbed (emitted) photon is close to the natural frequencies of a system of two non-interacting atoms. These processes have a clearly pronounced resonant character. Single-photon ionization in the case of remote collisions, accompanied by excitation transfer from one atom to another, was considered in Refs. 2 and 3, while two-photon ionization was considered in Ref. 4.

In this paper we consider processes that occur in atomic collisions and are accompanied by absorption of photons whose frequency differs from the natural atomic frequencies by an amount comparable with the frequencies itself. We shall call these processes nonreso-

nant.

We note that nonresonant excitation (detuning ~ 1000 cm^{-1}) was observed in recent experiments,⁵ as well as binding of atoms of alkali-metal vapor into a molecule following collisions in a laser-radiation field. If the detuning reaches a value on the order of the atomic frequency, then the nonresonant processes occur at close collisions, since the close approach can lead to cancellation of the detuning on account of the strong distortion of the energy levels of the interacting atoms (quasimolecule). For a consistent calculation of the cross sections in the adiabatic approximation it is therefore necessary, first, to know the exact parameters of the quasimolecule; second, to take into account the motion of the nuclei along the real rather than a straight-line trajectory. On the other hand, by measuring the cross sections of nonresonant processes in close collisions it is possible to obtain information on the parameters of the quasimolecule (the angular momenta of the transitions, the shapes of the potential curves).

In the next section of this paper we reduce the problem in the adiabatic approximation to a description, in