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Transitions Under the Influence of Local Interaction in Stochastic Motion

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Besides the case of linear intersecting terms (The Landau-Zener problem), interest attaches in the study of non-adiabatic transition to the case when both the terms and the interaction leading to the transition between them vary very rapidly near the intersection, and can be represented in the limit by δ-functions. The problem of calculating the transition probabilities in a two-level system for such local interactions and for stochastic motion along the term is solved for a rectangular interaction and Poisson process and for a δ-like potential and one-dimensional diffuse motion. The results are compared with those of the correlation theory.

A large group of kinetic effects can be interpreted with the aid of a time-dependent Hamiltonian $H = H[\sigma, q(t)]$, where σ represents the quantum variables, and $q(t)$ the classical trajectories regarded as functions of the time. The transition rates in such systems are usually calculated on the basis of the correlation theory[1], which is a variant of perturbation theory. In the present paper we solve the corresponding problem exactly for certain concrete cases, so that a comparison of our results with the data of the correlation theory makes it possible to determine more precisely the region of applicability of this theory.

For a condensed phase, each trajectory can be regarded, in many problems, as an individual realization of a certain random process $q(t)$ described by probability characteristics. We then obtain the following expression for the probabilities $W_{gg'}$ per unit time of the transitions between the states g and g' of the time-dependent Hamiltonian $H_0(\sigma)$ under the influence of a perturbation $V(\sigma, q)$ that varies randomly with time:

$$W_{gg'} = \frac{2}{\hbar^2}\operatorname{Re}\int_0^t d\tau \exp(i\omega_{gg'}\tau) \int dq_1 \int dq_2 V_{gg'}(\sigma, q_1) \times V_{g'g}(\sigma, q_2) P(q_2, \tau | q_1, 0) P(q_1), \tag{1}$$

where $P(q_2, t_2 | q_1, t_1)$ is the conditional probability of the random process $q(t)$, $P(q)$ is its one-dimensional distribution function, and $\hbar\omega_{gg'} = E_g - E_{g'}$. Disregarding the dependence of the matrix elements on the indices g and g', which is immaterial for our purpose, and putting $\omega_{gg'} = \omega_0$ and $V_{gg'} = {}^1\!/_2\hbar\omega_1(q)$, we rewrite (1) in the form

$$W = {}^1\!/_2 \operatorname{Re}\int_0^t d\tau\, e^{i\omega_0\tau} K(\tau), \tag{2}$$

where

$$K(\tau) = \langle \omega_1[q(0)] \omega_1^*[q(\tau)] \rangle$$

is the correlation function of the random quantity $\omega_1(q)$. If τ_c is the correlation time of the function $K(\tau)$ (defined as $\tau_c = K^{-1}(0) \int_0 |K(\tau)|\, d\tau$), then the condition for the applicability of formula (2) is usually written in the form

$$\langle \omega_1^2 \rangle \ll \max(\omega_0^2, \omega_c^2), \quad \omega_c = \tau_c^{-1}. \tag{3}$$

This condition presupposes that those trajectories $q(t)$ for which perturbation theory does not hold make a small contribution to W in comparison with the main (macroscopically overwhelming) mass of the trajectories.

It is easily seen that the condition (3) can be satisfied in such a situation only for nonlocal interactions $\omega_1(q)$, i.e., when the transitions occur at practically arbitrary values of q. Indeed, satisfaction of inequality (3) in this case allows us to state that perturbation theory does not hold only for trajectories for which the time dependence of $q(t)$ is such that $\omega_1[q(t)] \sim \exp(i\omega_0 t)$. But the measure of such trajectories is vanishingly small, and their contribution to W can be disregarded. In other words, the condition (3) is in fact a sufficient condition for the applicability of (2) in the case when the form of the function $\omega_1(q)$ and the character of the random process $q(t)$ are such that we have in order of magnitude $(\omega_1^2)_{\max} \sim \langle \omega_1^2 \rangle$. Such a case is realized for example in the calculation of the probabilities of relaxation processes between Zeeman nuclear magnetic energy levels under the influence of the dipole-dipole interaction of the magnetic moments of nuclei of one and the same molecule, modulated by rotational diffusion ("internal relaxation"). However, if $\omega_1(q)$ differs noticeably from zero only in a small region Δq, as is the case in collisions between two interacting particles, then $\langle \omega_1^2 \rangle$ can be arbitrarily small at sufficiently low concentrations. For trajectories that lead to transitions (and correspond to collisions), perturbation theory may indeed not be valid for the calculation of W. Since trajectories for which there are no collisions do not lead to transitions at all (although their measure is large), formula (2) may turn out to be incorrect in general.

We consider in this paper the calculation of the probabilities of the transitions under the influence of such local interactions. In particular, we derive the conditions under which expression (2) is valid in this case. We choose as a model the two-level Hamiltonian

$$H = H_0 + V, \quad H_0 = {}^1\!/_2\hbar[\omega_0 + \omega(q)]\sigma_z, \quad V = {}^1\!/_2\hbar\omega_1(q)\sigma_x, \tag{4}$$

where σ_x and σ_z are Pauli matrices. Without loss of generality, we can put

$$\langle \omega(q) \rangle = \langle \omega_1(q) \rangle = 0. \tag{5}$$

Indeed, if the condition (5) is not satisfied, then, by subtracting from ω and ω_1 their mean values and adding the corresponding terms to the constant part of the Hamiltonian, we can again obtain a Hamiltonian of the type (4) and condition (5) by rotating the coordinate system. Just as in our preceding paper[2], we have included in the unperturbed Hamiltonian H_0 the "secular" part of the time-dependent perturbation.

Since $q = q(t)$ is a random process, the transition probability W should be obtained by averaging, over all the possible realization of this process, the functional $W[q(t)]$ corresponding to the transition probability for the given realization. An expression for $W[q(t)]$ can be obtained if it is possible to solve the time dependent Schrodinger equation with a Hamiltonian (4) in which $q(t)$ is an arbitrary function of the time. This can be done exactly when $H_0 = 0$ and $H = (1/2)\hbar\omega_1(q)\sigma_x$, or else by regarding V as a perturbation. The first situation corresponds to a transition between degenerate states under the influence of the interaction produced upon collision of the particles. Let $\psi^{(1,2)}$ be the eigenfunctions of the operator σ_z. Then, if $\psi^{(1)} = 1$ and $\psi^{(2)} = 0$ at $t = 0$, we get at the instant of time t

$$\psi^{(2)} = \sin\left\{{}^1/_2\int_0^t \omega_1[q(t')]dt'\right\}, \tag{6}$$

and for the transition probability at the instant t we have

$$W_t[q(t)] = |\sin {}^1/_2\int_0^t \omega_1(q(t'))dt'|^2 = {}^1/_2\left[1 - \operatorname{Re}\exp\left\{i\int_0^t \omega_1(q(t'))dt'\right\}\right]. \tag{7}$$

The transition probability at the instant t, averaged over all the realizations, is

$$\begin{gathered} W_t = \langle W_t[q]\rangle = {}^1/_2\operatorname{Re}[1 - \varphi(t)], \\ \varphi(t) = \left\langle \exp\left[i\int_0^t \omega_1(q(t'))dt'\right]\right\rangle. \end{gathered} \tag{8}$$

Formula (8) represents averaging, over all the trajectories, of the exact expression for the probability of the transition from the state $\psi^{(1)}$ to the state $\psi^{(2)}$, and therefore takes into account automatically both direct and inverse transitions. The number of transitions per unit time can be obtained with the aid of (8) by normalizing the stationary random process $q(t)$ to the density of the particle flux through the point $q = q_0$ (corresponding to collision). We can expect W_t to become proportional to the time after a transient time interval (following the turning-on of the interaction at $t = 0$), i.e., at sufficiently long t, for under these conditions the "supply" of particles to the point q_0, where the interaction takes place, is constant, and the transition probability per interaction act does not depend on the time.

If the interaction V can be regarded as a small perturbation, then the transition probability obtained from perturbation theory does not take inverse transitions into account. Therefore, averaging this quantity over the trajectories, we find that it is proportional to the time for the stationary random process $q(t)$ normalized to the particle concentration. Indeed, in first order of perturbation theory we obtain for $\psi^{(2)}$ ($\psi^{(1)} = 1$ at $t = 0$)

$$\psi^{(2)} = i\int_0^t \omega_1(q(t_1))\exp\left\{i\int_{t_0}^{t_1} [\omega_0 + \omega(q(t'))]dt'\right\}dt_1,$$

and for the transition probability at the instant t, averaged over all trajectories, we get

$$W_t = \int_0^t dt_1\int_0^t dt_2\left\langle \omega_1(q(t_1))\omega_1(q(t_2))\exp\left\{i\int_{t_1}^{t_2} [\omega_0 + \omega(q(t'))]dt'\right\}\right\rangle$$

$$= \int_0^t dt_1\int_0^t dt_2\Phi(t_1, t_2). \tag{9}$$

For stationary Markov random processes, Φ depends only on the time difference:

$$\Phi(t_1, t_2) = \Phi(t_2 - t_1) = \Phi(\tau).$$

In addition, $\Phi(\tau) = \Phi^*(-\tau)$, and when $|\tau| \gg \tau_c$ (τ_c is the correlation time of the random quantities $\omega(q)$ and $\omega_1(q)$) we have $\Phi(\tau) \to 0$. Making in (9) the change of variables $\tau_2 - \tau_1 = \tau$ and $t_1 + t_2 = \xi$ and assuming that $t \gg \tau_c$, we find that $W_t = Wt$, and the transition probability per unit time is

$$W = 2\operatorname{Re}\int_0^\infty \Phi(\tau)d\tau. \tag{10}$$

The functions $\varphi(t)$ and $\Phi(t)$, in the general case of Markov stationary processes $q(t)$, can be calculated by introducing the quantity $Q(q_2, t_2 - t_1; q_1)$, which is the incomplete mean value of $\exp[i\int_{t_1}^{t_2}\omega(q)dt']$:

$$\begin{aligned} Q(q_2, t_2 - t_1; q_1) = \int dq^{(1)}\ldots dq^{(n)}P(q_1, t_1|q^{(1)}, t^{(1)})P(q^{(1)}, t^{(1)}|q^{(2)}, t^{(2)}) \\ \times\ldots P(q^{(n)}, t^{(n)}|q_2, t_2)\exp[i\omega(q^{(1)})(t^{(1)} - t_1) + i\omega(q^{(2)})(t^{(2)} - t^{(1)}) + \ldots \\ \ldots + i\omega(q_2)(t_2 - t^{(n)})]. \end{aligned} \tag{11}$$

In the calculation of φ and Φ, the quantity $\omega(q)$ in the exponential should be replaced by $\omega_1(q)$ and $\omega_0 + \omega(q)$, respectively. With the aid of Q we can represent the functions $\varphi(\tau)$ and $\Phi(\tau)$ in the form

$$\varphi(\tau) = \int dq_1dq_2Q(q_2, \tau; q_1)P(q_1), \tag{12}$$

$$\Phi(\tau) = \int dq_1\, dq_2\omega_1(q_1)\omega_1(q_2)Q(q_2, \tau; q_1)P(q_1). \tag{13}$$

If the conditional probability $P(q_2, t_2| q_1, t_1)$ satisfies a Kolmogorov equation of the type

$$\partial P / \partial t_2 = \mathscr{L}_{q_2}P, \tag{14}$$

then we can obtain for Q[3,4]

$$\partial Q / \partial t_2 = i\omega(q_2)Q + \mathscr{L}_{q_2}Q \tag{15}$$

with an initial condition

$$Q(q_2, 0; q_1) = \delta(q_2 - q_1). \tag{16}$$

We note that Eq. (15) is equivalent to an integral equation whose solution satisfies the initial condition, namely

$$Q(q_2, \tau; q_1) = P(q_2, \tau|q_1, 0) + i\int_0^\tau dt'\int dq'P(q_2, \tau|q', t')\omega(q')Q(q', t'; q_1). \tag{17}$$

We choose the one-dimensional Poisson process as the model of random motion for transitions between nondegenerate states $\omega_0 + \omega(q) = 0$ under the influence of a local interaction. We assume that the trajectory $q(t)$ can be represented by discrete jumps through a fixed distance a, so that at each instant of time $q = na$, where $n = 0, \pm1, \pm2, \ldots$ The jumps occur at an average frequency ν and furthermore in only one direction, so that in each jump q changes from $q = na$ to $q = (n + 1)a$. Let the transition-inducing interaction $\tilde{\omega}_1(q)$ differ from zero only for the position $q = an_0$, where it is equal to Ω. For $\tilde{\omega}_1(q) = \tilde{\omega}_1(n)$ we can write

$$\tilde{\omega}_1(n) = \Omega\delta_{nn_0}. \tag{18}$$

According to the foregoing, in order to satisfy condition (5) we turn on in the operator H_0, by means of a renormalization, the part $\tilde{\omega}_1$, which does not vanish upon averaging (see (16). Then the quantity $\omega_1(q) = \omega_1(n)$ in (6) takes the form

$$\omega_1(n) = \Omega\delta_{nn_0} - P(n_0)\Omega, \tag{19}$$

where P(n) is the probability of being located in the position n. If the particle flux through the point q = na is denoted by f, then we get for P(n)

$$P(n) = P_0 = f/\nu, \tag{20}$$

where P_0 is the concentration. Formula (12) takes the form

$$\varphi(\tau) = P_0 \sum_{n_1,n_2} Q(n_2, \tau; n_1). \tag{21}$$

Recognizing that the conditional probability $P(n_2, t_2 | n_1, t_1)$ for a Poisson process is[5]

$$P(n_2,\ t_2|n_1,\ t_1) = [\nu(t_2 - t_1)]^{n_2-n_1} e^{-\nu(t_2-t_1)} / (n_2 - n_1)!, \tag{22}$$

and representing (17) in the form

$$Q(n_2, \tau; n_1) = P(n_2, \tau | n_1, 0)\exp(iP_0\Omega\tau) + i\Omega\int_0^\tau P(n_2, \tau | n_0, t') \times \exp[iP_0\Omega(\tau - t')]Q(n_0, t'; n_1)\,dt', \tag{23}$$

we obtain

$$\varphi(\tau) = \exp(iP_0\Omega\tau)\left[1 + i\Omega P_0 \int_0^\tau \frac{\nu - i\Omega e^{i\Omega t - \nu t}}{\nu - i\Omega} dt\right]. \tag{24}$$

Substituting this result in (8) we find that for an interval that is large in comparison with the average time of an individual jump but is small in comparison with the average interaction frequency (small concentrations), i.e., at

$$\nu\tau \gg 1, \quad P_0\Omega\tau \ll 1, \tag{25}$$

the quantity W_T is proportional to the time, so that

$$W = \frac{1}{2} P_0 \frac{\Omega^2\nu}{\nu^2 + \Omega^2}. \tag{26}$$

Conditions (25) can be satisfied simultaneously when $\nu \gg P_0\Omega$ and $\nu^2 \gg f\Omega$, i.e., in the case of sufficiently small fluxes. When $\Omega \ll \nu$, the result (26) goes over into the well known result of correlation theory[1], $W = \frac{1}{2}P_0\Omega^2\nu^{-1}$, with $P_0\Omega^2 = \langle \omega_1^2 \rangle$, $\tau_c = \nu^{-1}$. We see therefore that the condition for the applicability of correlation theory is not the satisfaction of the conditions (3) but the satisfaction of the inequality $\nu \gg \Omega$, meaning that for local interactions perturbation theory holds only when the correlation frequency of the random process is much higher than the characteristic interaction frequency Ω at the instant of "collision." Thus, the mean value of the interaction $\langle \omega_1^2 \rangle$ can be arbitrarily small (at low concentrations), but the results of the correlation theory turn out to be incorrect if $\Omega \gtrsim \nu$ (the transition probability per "collision" calculated from the correlation theory may turn out to be larger than 1).

We now consider the case when the interaction in (6) can be regarded as a perturbation. We note by way of an introduction that the formula (10) holds for the transition probability not because the local interaction is small at the instant of encounter (during the "collision") but only because the non-secular part of this interaction is small, since the corresponding secular part is taken into account by formula (10) exactly. For the case $\omega_0\tau_c \gg 1$, similar problems were considered by Karamyan and one of the authors[6]; the present analysis is suitable for arbitrary τ_c. Let us calculate the function $\Phi(\tau)$ for a Poisson process, when

$$\omega_1 = \Omega_1\delta_{nn_0} - \Omega_1 P_0, \quad \omega = \Omega\delta_{nn_0} - \Omega P_0.$$

Formula (13) yields in this case

$$\Phi(\tau) = P_0\Omega_1^2\Big\{Q(n_0, \tau; n_0) - P_0 \sum_n [Q(n, \tau; n_0) + Q(n_0, \tau; n)] + P_0^2 \sum_{n,n'} Q(n, \tau; n')\Big\}. \tag{27}$$

At small concentrations it is necessary to take into account only the first term in the curly brackets. Indeed, as seen from (11), a majorizing estimate of $Q(n_2, \tau; n_1)$ is

$$|Q(n_2,\ \tau;\ n_1)| \leqslant P(n_2,\ \tau | n_1,\ 0).$$

Therefore $|\ \Sigma Q(n', \tau; n)| \leq 1$ and at small P_0 we can assume for Φ:

$$\Phi(\tau) = P_0\Omega_1^2 Q(n_0,\ \tau;\ n_0). \tag{28}$$

We note that a nonzero value of P_0 corresponds (at a finite number of particles) to a process that is limited in the space $-N \leq n < N$, so that the quantities, P_0, and Q in (27) should be tagged with the index N. Actually, we take the limit as $N \to \infty$, confining ourselves to terms linear in the concentration.

As seen from (9), the quantity $\omega(q)$ in (15) for a stepwise local interaction is equal to

$$\omega_0 + \omega(n) = \omega_0 + \Omega\delta_{nn_0}.$$

Therefore the substitution

$$Q(n',\ \tau;\ n) = \exp(i\omega_0\tau)Q'(n',\ \tau;\ n)$$

leads to Eq. (17) for Q'. From formula (22) we obtain for $Q'(n_0, \tau; n_0)$

$$Q'(n_0, \tau; n_0) = e^{-\nu\tau} + i\Omega\int_0^\tau e^{-\nu(\tau-t)}Q'(n_0, t; n_0)\,dt,$$

whence

$$Q'(n_0, \tau; n_0) = e^{-\nu\tau + i\Omega\tau}, \qquad \Phi(\tau) = P_0\Omega_1^2 e^{-\nu\tau + i\Omega\tau + i\omega_0\tau}.$$

From (10) we obtain directly the following expression for the transition probability per unit time between states with definite σ_z under the influence of $\omega_1(n) = \Omega_1[\delta_{nn_0} - P_0]$:

$$W = 2P_0\Omega_1^2 \frac{\nu}{\nu^2 + (\Omega + \omega_0)^2}, \tag{29}$$

whereas the correlation theory yields in this case

$$W = 2P_0\Omega_1^2 \frac{\nu}{\nu^2 + \omega_0^2}. \tag{30}$$

The results (29) and (30) coincide if $\Omega \ll \omega_0$, which is the condition for the applicability of the correlation theory, whereas the sufficient condition for the validity of (29) is

$$\Omega_1^2 \leqslant \nu^2 + (\Omega + \omega_0)^2 \tag{31}$$

(the probability of a transition in the average time of one "collision" is much less than 1). Expression (29) has a maximum at $\Omega = -\omega_0$; the physical meaning of this is obvious, since $\Omega = -\omega_0$ corresponds to the resonance conditions at the instant of collision.

We now consider the case when

$$\omega_1(q) = \gamma_1[\delta(q) - P_0], \quad \omega(q) = \gamma\delta(q)$$

(P_0 has now the dimension of reciprocal length) and $q = q(t)$ is a random process whose conditional probability satisfies the diffusion equation

$$\partial P(q_2, t|q_1, 0) / \partial t = D\partial^2 P(q_2, t|q_1, 0) / \partial q_2^2.$$

In analogy with (28) we obtain for $\Phi(t)$

$$\Phi(t) = P_0\gamma_1^2 Q(0, t; 0). \tag{32}$$

To calculate $Q(0, t; 0)$ we can again use (17). We shall describe, however, another and more general method which is convenient not only for local interactions.

Equation (15) and the initial condition (16) at $\omega(q) = \gamma\delta(q)$ are satisfied by the function

$$Q(q_2, \tau; q_1) = e^{i\omega_0\tau} \sum_k e^{-k^2D\tau} y_k(q_1)\tilde{y}_k^*(q_2), \tag{33}$$

where y_k is an eigenfunction of the operator $\widetilde{\mathscr{L}}_q = \partial^2/\partial q^2 - 2ib\delta(q)$, corresponding to the eigenvalue $-k^2$ ($b = \gamma/2D$),

$$\mathscr{L}_q y_k = -k^2 y_k, \tag{34}$$

and $\tilde{y}_k$ is the eigenfunction of the conjugate operator $\widetilde{\mathscr{T}}_q = \partial^2/\partial q^2 - 2ib\delta(q)$. The functions y_k and $\tilde{y}_k$ were chosen to satisfy the equation

$$\int y_k(q)\tilde{y}_{k'}^*(q)\,dq = \delta_{kk'}. \tag{35}$$

If the diffusion takes place on an infinite straight line, then the sum in (33) goes over into an integral, the Kronecker symbol in (35) becomes a δ-function; the boundary condition in this case is that the solution be bounded on the entire line. For diffusion in the region $-L \le q \le L$, the boundary conditions correspond to absence of a probability-density flux through the boundary

$$\left.\frac{\partial y}{\partial q}\right|_{q=L} = \left.\frac{\partial y}{\partial q}\right|_{q=-L} = 0. \tag{36}$$

The bounded solution of (34) is

$$y_k(q) = a_k(\cos kq - ibk^{-1}\sin k|q|), \tag{37a}$$

and the solution of the conjugate equation yields

$$\tilde{y}_k(q) = \tilde{a}_k(\cos kq + ibk^{-1}\sin k|q|), \tag{37b}$$

with k real (the solution $y_k = a_k' \sin kq$ is of no interest since (32) contains only $y_k(0)$). It is easy to verify directly that the condition (35) is satisfied if we put

$$a_k\tilde{a}_k^* = \frac{1}{\pi}\frac{k^2}{k^2 - b^2}. \tag{38}$$

Let the contour C coincide with the real k axis everywhere except at the points $k_{1,2}^2 = \pm b$; the points $\pm b$ are circled by infinitesimally small semicircles from above and below, respectively. We then have for Q

$$Q(q_2, t; q_1) = \frac{1}{\pi}\int_C \frac{k^2\exp(-k^2\gamma t + i\omega_0 t)}{k^2 - b^2} \times [\cos kq_1 - ibk^{-1}\sin k|q_1|][\cos kq_2 - ibk^{-1}\sin k|q_2|]\,dk. \tag{39}$$

It is easy to verify that when the indicated integration contour is used the functions y_k and $\tilde{y}_k$ determined from (37) and (38) satisfy the relation

$$\int_C y_k(q_1)\tilde{y}_k^*(q_2)\,dk = \delta(q_1 - q_2),$$

which ensures satisfaction of the initial condition (16). We note that we obtained the contour C by considering diffusion on a bounded segment $-L \le q \le L$ (i.e., by using the boundary condition (36) and normalizing the solution in accordance with (35); we then cbtain $a_k\tilde{a}_k^* = k^2[(k^2 - b^2) \times L - ib]^{-1}$) and taking the limit as $L \to \infty$.

Formulas (10), (32), and (39) yield

$$W = \frac{2\gamma_1^2}{\pi}\operatorname{Re}\int_C \frac{k^2\,dk}{(k^2 - b^2)(k^2D - i\omega_0)}$$

$$= \frac{P_0\gamma_1^2}{D}\left(\frac{|\omega|}{2D}\right)^{1/2}\begin{cases}\left[b^2 + b\left(\frac{2|\omega|}{D}\right)^{1/2} + \frac{|\omega|}{D}\right]^{-1}, & \omega_1 > 0\\ \left[b^2 - b\left(\frac{2|\omega|}{D}\right)^{1/2} + \frac{|\omega|}{D}\right]^{-1}, & \omega_1 < 0\end{cases}. \tag{40}$$

Under no condition does expression (40) for the transition probability go over into the corresponding formula obtained for the probability from correlation theory. This is understandable if it is recalled that the local interaction was simulated by a δ-function, and consequently a condition such as (31) can never be satisfied.

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