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► **To cite this version:**

V. Čápek. Symmetry breaking in the double-well model: ohmic dissipation with unrelaxed (Feynman-Vernon) initial conditions. *Journal de Physique*, 1989, 50 (7), pp.775-781. 10.1051/jphys:01989005007077500 . jpa-00210955

HAL Id: jpa-00210955

<https://hal.science/jpa-00210955>

Submitted on 4 Feb 2008

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Classification

Physics Abstracts

74.50 — 05.30 — 05.40

Symmetry breaking in the double-well model : ohmic dissipation with unrelaxed (Feynman-Vernon) initial conditions

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(Reçu le 1^{er} juin 1988, accepté sous forme définitive le 22 novembre 1988)

Résumé. — Nous formulons le problème de la brisure de symétrie aux temps grands d'une particule dans un double puits interagissant avec des phonons dans le cas d'un modèle minimal, standard, à deux sites, avec une distribution initiale asymétrique de la particule et des phonons thermiques, mais non relaxés. Nous traitons explicitement l'équation maîtresse généralisée dans l'approximation de Born. L'extrapolation de la solution aux temps infinis conduit à des critères pour la brisure de symétrie asymptotique en accord avec ceux connus pour les conditions initiales relaxées.

Abstract. — The problem of asymptotic-time symmetry breaking for a particle in a symmetric double-well, interacting with phonons, is formulated for a standard minimal two-site model and for initially asymmetric particle distribution and thermal but unrelaxed phonons. The time-convolutionless Generalized Master Equations can be handled explicitly in the Born approximation. Formally extending the solution to the infinite time, the criteria for the asymptotic-time symmetry breaking comply with those known for the relaxed initial condition.

1. Introduction.

In 1982, Chakravarty [1] and Bray with Moore [2] first treated the so called double-well model for a particle interacting with the thermodynamic bath (phonons henceforth). Possible reduction to the two-site model under appropriate conditions, relative simplicity and, simultaneously, sufficient generality to describe simultaneous complementary processes like transfer (diffusion) and relaxation stimulated increasing interest in the model [3-12]. Its drawback is, however, that a full exact solution under sufficiently general conditions will probably never be found. Therefore, we are forced to resort to approximations.

Already Bray with Moore [2] turned our attention to the fact that, imposing asymmetric initial conditions even in a fully symmetric two-site model, one might get still an asymmetric particle distribution in the infinite-time limit under specific conditions provided that the interaction with phonons is sufficiently strong and the initial phonon temperature is zero.

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There is a question, however, whether approximate treatments can be used to discuss the dynamics at arbitrarily long times or not. Nevertheless, calculations by Aslangul, Pottier and Saint-James [13] (and more generally [14]) based on second-order approximate time-convolutionless Generalized Master Equations confirmed the standing opinion. This means that for the symmetric model, so called Ohmic coupling with phonons with the coupling parameter $\alpha \leq 1$ (for these notions see [12] or below) and temperature $T = 0$, no asymptotic-time symmetry breaking exists. For $\alpha > 1$, such a symmetry breaking takes place. Nevertheless, the asymptotic asymmetry is less than that in the initially asymmetric initial condition. For $T > 0$, no symmetry breaking in the asymptotic time domain is believed to exist for the Ohmic type of interaction with phonons. An analogous treatment for the subohmic and superohmic cases (not treated in this work) may be found in [15].

In view of the importance of these results, it is desirable to check the conclusions by discussing if they were (or were not) influenced by the

- a) initial conditions used,
- b) approximate treatments of the kinetics, and
- c) model Hamiltonian itself.

Following this program, in this work, we should like to discuss the first point of the list. As usual, in [13-14], the relaxed initial conditions were used. This means that if the particle is initially in e.g. the left-hand-side state, the phonons (interacting with it) are initially assumed to be fully accommodated to its presence and position. This results in such effects as a full formation of the polaron cloud and the shift of the mean energy (polaron shift) before the motion starts. Instead, in this work, we choose initially unrelaxed phonons. Locating the particle at $t = 0$ in e.g. the left-hand-side state leads to its motion (possible transfer to the right-hand-side state) accompanied simultaneously with a gradual formation of the polaron cloud. This causes technical problems which can be fortunately solved. In order to keep the connection with [13-14], we also use the time-convolutionless Generalized Master Equations in the second order (in the particle-hopping term) approximation. In order to avoid the appearance of the initial condition term, however, the usual small-polaron transformation of the Hamiltonian must be avoided, i.e. we work in the unrelaxed basis. Owing to the different initial condition, one cannot expect the same solution (including its formal limit $t \rightarrow +\infty$). Nevertheless, in order to be able to ascribe a physical meaning to the usual criteria for the appearance of the asymptotic symmetry breaking, these criteria should result the same. This is really confirmed below. For simplicity, we assume $T = 0$ only where T designates the initial phonon temperature. The case of $T \neq 0$ will be shortly mentioned in the appendix.

2. Model and formalism.

Let us start with the Hamiltonian of the symmetric two-site double-well model (spin-boson Hamiltonian)

$$\begin{aligned}
 H &= -\frac{1}{2} \hbar \omega_0 \sigma_x + \sum_{i=1}^M \hbar G_i (b_i + b_i^\dagger) \sigma_z + \sum_{i=1}^M \hbar \omega_i b_i^\dagger b_i \\
 &\equiv -\frac{1}{2} \hbar \omega_0 (a_1^\dagger a_2 + a_2^\dagger a_1) + \sum_{i=1}^M \hbar G_i (b_i + b_i^\dagger) (a_1^\dagger a_1 - a_2^\dagger a_2) + \sum_{i=1}^M \hbar \omega_i b_i^\dagger b_i \\
 &\equiv \mathcal{K} + H_{\text{int}} + H_{\text{ph}} .
 \end{aligned} \tag{1}$$

Here $b_i (b_i^\dagger)$ and $a_j (a_j^\dagger)$ are the phonon annihilation (creation) operators and annihilation (creation) operators of the particle in the respective state ($j = 1$ and 2 for the left- and right-hand site, respectively). For the single particle used, it is not important whether it is a fermion

or boson. ω_i are the phonon frequencies while ω_0 gives the separation between single-particle eigenenergies and, simultaneously, the frequency of the coherent particle oscillations $1 \leftrightarrow 2$ if the coupling constants G_i were zero.

Let us now specify the initial condition for the full density matrix $\rho_F(t)$. In accordance with what has been said above, we put

$$\rho_F(0) = \rho_S(0) \otimes \rho_B(0), \tag{2a}$$

$$\rho_S(t) = \text{Tr}_B \rho_F(t), \quad \rho_B(t) = \text{Tr}_S \rho_F(t) \tag{2b}$$

where the indices B and S designate the bath (phonons) and system (particle), respectively. This means (in contrast to e.g. [13-14]) that the bath is prepared (at $t \leq 0$) in a thermal state, being decoupled from the system. The coupling to the system (particle) is switched on at $t = 0$ simultaneously with the possibility for the particle to move (Feynman-Vernon initial conditions [16]). In [13-14], during preparation of the initial state, the bath is assumed to be interacting with the particle. At $t = 0$, just the possibility for the particle to move is switched on in [13-14], in contrast with our treatment here.

For the projection superoperator (an operator in the Liouville space) D , we take the Argyres and Kelley projector

$$DA = \text{Tr}_B (A) \otimes \rho_B(0). \tag{3}$$

Here A is an arbitrary operator in the Hilbert space of the particle with phonons. Because of (2a),

$$(1 - D) \rho_F(0) = 0. \tag{4}$$

Therefore, in the Shibata, Hashitsume, Takahashi and Shingu identity [17-18] resulting from the Liouville equation

$$\begin{aligned} \frac{\partial}{\partial t} D \tilde{\rho}_F(t) = & -iD\tilde{\mathcal{L}}(t) \left[1 + i \int_0^t \exp_{\leftarrow} \left\{ -i \int_{\tau_1}^t (1 - D) \tilde{\mathcal{L}}(\tau_2) d\tau_2 \right\} \times \right. \\ & \times (1 - D) \tilde{\mathcal{L}}(\tau_1) D \exp_{\rightarrow} \left. \left\{ i \int_{\tau_1}^t \tilde{\mathcal{L}}(\tau_3) d\tau_3 \right\} d\tau_1 \right]^{-1} \\ & \times \left[\exp_{\leftarrow} \left\{ -i \int_0^t (1 - D) \tilde{\mathcal{L}}(\tau) d\tau \right\} (1 - D) \rho_F(0) + D \tilde{\rho}_F(t) \right], \tag{5} \end{aligned}$$

the initial condition term $\sim (1 - D) \rho_F(0)$ can be omitted. In (5), for any A

$$\begin{aligned} \tilde{A}(t) = & e^{\frac{i}{\hbar} H_0 t} A e^{-\frac{i}{\hbar} H_0 t} \equiv e^{iL_0 t} A, \\ L_0 = & \frac{1}{\hbar} [H_0, \dots], \quad H_0 = H_{\text{int}} + H_{\text{ph}} \tag{6} \end{aligned}$$

designates the operator A in the interaction picture ; \mathcal{L} is given as

$$\mathcal{L} \equiv \frac{1}{\hbar} [\mathcal{H}, \dots] \sim \omega_0 \tag{7}$$

so that

$$\begin{aligned} \tilde{\mathcal{L}}(t) \tilde{A}(t) \equiv \tilde{\mathcal{L}}\tilde{A}(t) = & \frac{1}{\hbar} [\tilde{\mathcal{H}}(t), \tilde{A}(t)] \\ = & e^{iL_0 t} \mathcal{L} e^{-iL_0 t} \tilde{A}(t) \sim \omega_0. \tag{8} \end{aligned}$$

Following the same line of reasoning as in [13-14], we approximate (5) (taking (4) into account) as

$$\frac{\partial}{\partial t} D \tilde{\rho}_F(t) = -i D \tilde{\mathcal{L}}(t) D \tilde{\rho}_F(t) - D \tilde{\mathcal{L}}(t) (1 - D) \int_0^t \tilde{\mathcal{L}}(\tau) d\tau D \tilde{\rho}_F(t). \quad (9)$$

In order to obtain (9) from (5), we have omitted the exponentials in (5) and approximated $[1+x]^{-1} \approx 1-x$, $x \sim \tilde{\mathcal{L}} \sim \omega_0$. Equation (9) is formally exact to the second order in ω_0 . Right here, however, we should like to point out that conditions for the validity of this second-order (Born) approximation may be well violated for arbitrarily small but finite ω_0 as far as the time t is increased beyond any limit. This is clearly seen from the identity

$$i \int_0^t \exp_{\leftarrow} \left\{ -i \int_{\tau_1}^t (1-D) \tilde{\mathcal{L}}(\tau_2) d\tau_2 \right\} (1-D) \tilde{\mathcal{L}}(\tau_1) D \exp_{\rightarrow} \left\{ i \int_{\tau_1}^t \tilde{\mathcal{L}}(\tau_3) d\tau_3 \right\} d\tau_1 = \\ = -1 + D + \exp_{\leftarrow} \left\{ -i \int_0^t (1-D) \tilde{\mathcal{L}}(\tau_2) d\tau_2 \right\} (1-D) \exp_{\rightarrow} \left\{ i \int_0^t \tilde{\mathcal{L}}(\tau_3) d\tau_3 \right\}. \quad (10)$$

The left-hand side of (10) appears in (5) and its formal proportionality to ω_0 is relevant for the expansion arguments. On the other hand, the right-hand side of (10) is not (for sufficiently high t) of the order $\sim \omega_0$ any more. This observation makes our point b) in the above programme meaningful. We shall return to this point in a next publication (see also [15] for a comment in this respect). Here, it is worth mentioning that the Kasner theory as applied to the present problem [19] is also formally exact to the second order in ω_0 . Nevertheless, for a symmetric double-well, the result of [19] in the asymptotic time-domain does not fully agree with the standard treatment. This fact may be also ascribed to an uncertain validity of any second-order theory in the long-time limit.

3. Kinetic equations.

Designating

$$\rho_{ij}(t) = (\text{Tr}_B \tilde{\rho}(t))_{ij}, \quad i, j = 1, 2, \quad (11)$$

equation (9) may be written as

$$\frac{\partial}{\partial t} \rho_{ij}(t) = \sum_{k,l=1}^2 B_{ijkl}(t) \rho_{kl}(t), \quad i, j = 1, 2. \quad (12a)$$

Before specifying the coefficients, we should like to point out that $\rho_{ij}(t)$ are the matrix elements of the reduced (particle-) density matrix not in the Schrödinger but in the interaction picture. Therefore,

$$\rho_{lm}(t) = \sum_{\lambda} (\tilde{\rho}(t))_{l\lambda, m\lambda} = \sum_{\lambda} \left(e^{\frac{i}{\hbar} H_0 t} \rho_F(t) e^{-\frac{i}{\hbar} H_0 t} \right)_{l\lambda, m\lambda} \\ = \sum_{\lambda} e^{\frac{i}{\hbar} (E_{l\lambda}^0 - E_{m\lambda}^0) t} (\rho_F(t))_{l\lambda, m\lambda}. \quad (13)$$

Here $E_{l\lambda}^0$ are eigenenergies corresponding to eigenstates $l\lambda$ of H_0 with the particle in state l

and with the phonon state (here relaxed around the particle) λ . Therefore, the diagonal elements

$$\rho_{ll}(t) = \sum_{\lambda} (\rho_F(t))_{l\lambda, l\lambda} = (\rho_S(t))_{ll} \tag{14}$$

which may be calculated from (12a), retain the physical significance of probabilities of finding the particle in the left ($l = 1$) or the right ($l = 2$) state.

In (12a), coefficients $B_{ijkl}(t)$ are quite complicated in general. Designating states of the bath by Greek indices, it is from (9)

$$B_{ijkl}(t) = -i \sum_{\lambda\mu\nu} \left(\tilde{\Gamma}(t) - \tilde{\Gamma}(t)(1 - D) \int_0^t \tilde{\Gamma}(\tau) d\tau \right)_{i\lambda, j\lambda, k\mu, l\nu} (\rho_B(0))_{\mu\nu}. \tag{12b}$$

In order to obtain explicit formulae, we take as usual the case of the Ohmic dissipation with the exponential cut-off, in which (in the limit $M \rightarrow +\infty$)

$$\sum_{i=1}^M G_i^2 \delta(\omega - \omega_i) \approx G(\omega)^2 \rho(\omega) = \frac{\alpha}{2} \omega l^{-\omega/\omega_C}. \tag{15}$$

Here, the cut-off frequency ω_C is believed to be unimportant for final conclusions when $k_B T \ll \hbar\omega_C$. We set $T = 0$; then the explicit formulae for $B_{ijkl}(t)$ simplify so that

$$\begin{aligned} B_{1111}(t) &= -B_{1122}(t) = -B_{2211}(t) = B_{2222}(t) = B_{1212}(t) = B_{2121}(t) = \\ &= -\frac{1}{2} \omega_0^2 \int_0^t d\tau [\exp \{-\alpha \ln [1 + \omega_C^2(t - \tau)^2]\} \times \\ &\quad \times \cos \{2\alpha [\arctg \omega_C(t - \tau) + \arctg \omega_C t - \arctg \omega_C \tau]\} - \\ &\quad - \exp \{-\alpha \ln ([1 + \omega_C^2 t^2][1 + \omega_C^2 \tau^2])\}], \end{aligned} \tag{16a}$$

$$\begin{aligned} B_{1112}(t) &= -B_{1121}(t) = -B_{2212}(t) = B_{2221}(t) \\ &= B_{1211}(t) = -B_{1222}(t) = -B_{2111}(t) = B_{2122}(t) \\ &= -\frac{i\omega_0}{2} \frac{1}{\{1 + \omega_C^2 t^2\}^\alpha}, \end{aligned} \tag{16b}$$

$$\begin{aligned} B_{1221}(t) = B_{2112}(t) &= -\frac{1}{2} \omega_0^2 \int_0^t d\tau \left[\exp \left\{ -\alpha \ln \frac{(1 + \omega_C^2 t^2)(1 + \omega_C^2 \tau^2)}{1 + \omega_C^2(t - \tau)^2} \right\} \times \right. \\ &\quad \times \cos \{2\alpha [\arctg \omega_C(t - \tau) + \arctg \omega_C t - \arctg \omega_C \tau]\} - \\ &\quad \left. - \exp \{-\alpha \ln ([1 + \omega_C^2 t^2][1 + \omega_C^2 \tau^2])\} \right]. \end{aligned} \tag{16c}$$

From (12) and (16a-c), we get a set of two equations

$$\begin{aligned} \frac{\partial}{\partial t} [\rho_{11}(t) - \rho_{22}(t)] &= 2 B_{1111}(t)[\rho_{11}(t) - \rho_{22}(t)] + 2 B_{1112}(t)[\rho_{12}(t) - \rho_{21}(t)], \\ \frac{\partial}{\partial t} [\rho_{12}(t) - \rho_{21}(t)] &= 2 B_{1112}(t)[\rho_{11}(t) - \rho_{22}(t)] + [B_{1111}(t) - B_{1221}(t)][\rho_{12}(t) - \rho_{21}(t)], \end{aligned} \tag{17}$$

which must be solved simultaneously in order to decide which is the asymptotic ($t \rightarrow +\infty$) value of $\rho_{11}(t) - \rho_{22}(t)$. The solution reads

$$\begin{bmatrix} \rho_{11}(t) - \rho_{22}(t) \\ \rho_{12}(t) - \rho_{21}(t) \end{bmatrix} = \exp \left\{ \int_{t_0}^t d\tau \begin{bmatrix} 2 B_{1111}(\tau), 2 B_{1112}(\tau) \\ 2 B_{1112}(\tau), B_{1111}(\tau) - B_{1221}(\tau) \end{bmatrix} \right\} \cdot \begin{bmatrix} \rho_{11}(t_0) - \rho_{22}(t_0) \\ \rho_{12}(t_0) - \rho_{21}(t_0) \end{bmatrix} \quad (18)$$

where $t_0 \geq 0$ is an arbitrary new time-origin.

4. Asymptotic symmetry breaking.

The problem of the asymptotic symmetry breaking now becomes simple : If the matrix integral in the exponential in (18) has both eigenvalues finite when $t \rightarrow +\infty$, a symmetry breaking occurs. The sign and relative magnitude of B -coefficients then ensure that $|\rho_{11}(+\infty) - \rho_{22}(+\infty)| < |\rho_{11}(t_0) - \rho_{22}(t_0)|$. If one of the eigenvalues remained finite and the second one turned to (minus) infinity, there would be the asymptotic symmetry breaking for almost all initial conditions. Finally, if both eigenvalues of the matrix-integral in the exponential in (18) turn to (minus) infinity when $t \rightarrow +\infty$, there is no asymptotic symmetry breaking. Other possibilities are excluded by the magnitudes and signs of B -coefficients in (16a-c) as well as by physical considerations.

It is clear that because of the arbitrary choice of t_0 , the question turns to the asymptotic behaviour of $B_{ijkl}(t)$ at high enough time-arguments. Let us therefore choose t_0 finite but as large that the asymptotic form of $B_{ijkl}(t)$ in (16a-c) can be used for all $t \geq t_0$. It is not difficult to see that beyond certain t_0 , $B_{1111}(t)$ dominates over $B_{1112}(t)$ as well as $B_{1221}(t)$ so that the eigenvalues of the matrix integral in the exponential in (18) are for $t \rightarrow +\infty$ simply $\approx 2 \int_{t_0}^{+\infty} B_{1111}(\tau) d\tau$ and $\approx \int_{t_0}^{+\infty} B_{1111}(\tau) d\tau$. Hence, from the above three possibilities, just the first and the third ones may take place. Because

$$\begin{aligned} B_{1111}(t) &\approx -\frac{1}{2} \frac{\omega_0^2}{\omega_C(1-2\alpha)} \operatorname{Re} [i(1-i\omega_C t)^{-2\alpha+1}], \quad t \rightarrow +\infty, \alpha \neq \frac{1}{2}, \\ B_{1111}(t) &\approx -\frac{1}{2} \frac{\omega_0^2}{\omega_C} \operatorname{Re} [i \ln(1-i\omega_C t)], \quad t \rightarrow +\infty, \alpha = \frac{1}{2}, \end{aligned} \quad (19)$$

one easily reveals that for $0 < \alpha \leq 1$, there is no symmetry breaking when $t \rightarrow +\infty$. Similarly, for $\alpha > 1$, there is the symmetry breaking in our model. Hence, the kinetic equation treatment which is formally exact to the second order in ω_0 , yields in our case of the unrelaxed initial condition the same criterion for the symmetry breaking as its counterpart for the relaxed initial condition at $T = 0$ [13]. This was a necessary condition for ascribing a physical meaning to this criterion.

Acknowledgment.

The author should like to thank the University of Stuttgart, namely Prof. M. Wagner, and the Max-Planck-Institut für Festkörperforschung, Stuttgart, namely Prof. O. K. Andersen, for their kind hospitality and support during his stay in the Federal Republic of Germany.

Appendix.

The treatment presented here may be easily extended to the case of $T > 0$. Nevertheless, in detail, it becomes more complicated. Qualitatively, all the coefficients $B_{ijkl}(t)$ are of the same form as in (16a-c), except for additional (approximately exponential) factors under the integrals (compare with [13]). Fortunately, we found that in the asymptotic time domain, the

question of their qualitative behaviour may be turned to that solved in [13]. In other words, e.g. $B_{1111}(t) \rightarrow \text{const} < 0$, $t \rightarrow +\infty$. Consequently, there is no asymptotic-time symmetry breaking for $T > 0$.

This result, though it is in full agreement with the usual opinion, deserves, however, a further discussion. It means that increasing T increases the effective probability with which the particle diffuses to the opposite site. On the other hand, Hamiltonian (1) describes no direct lowest-order phonon-assisted hopping processes which might be promoted by increasing T . On the contrary, mean value of the squared small-polaron overlap between states of the particle in sites 1 and 2 is known to decrease with increasing T . Hence, the case of $T > 0$ also deserves some further discussion from the point of view of higher-order processes omitted in standard lowest-order treatments.

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