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Dipole magic echo in thermodynamical systems

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Abstract. The irreversible evolution component of the subsystem of non-secular dipole-dipole interactions under conditions of time inversion is considered on the basis of the non-equilibrium thermodynamics methods. The abridged description of the spin system evolution is effected with the help of projection operators and the introduction of the temperatures of secular, non-secular and Zeeman subsystems. Consideration of cross-correlation attenuation during the emergence of the temperature in the sub-system leads to a good agreement with experiment.

1. Introduction

The problem of correlation between reversibility and irreversibility in the development of macrosystems is generally understood as the problem of describing irreversible processes on the basis of reversible equations of mechanics. Skrebnev *et al* (1987) and Skrebnev *et al* (1987) considered this problem from a different aspect, namely as a problem of correlation between reversibility and irreversibility in the development of the system under conditions of change in the time sign in the equations of motion. Observing the evolution of various terms of the spin-system density matrix in a situation corresponding to a change (with specified accuracy) in the time sign in the equations of motion we have the possibility of testing directly the applicability of reversible equations of mechanics for the description of the evolution of the macrosystem.

The basic possibility of posing the question of time-inversion experiments follows from the form of the solution of the Liouville equation:

$$\sigma(t) = \exp(-i\mathcal{H}t)\sigma(0)\exp(i\mathcal{H}t). \quad (1)$$

The sign change in the Hamiltonian in (1) is fully equivalent to the change in time sign. The development in the 1960s of the high resolution NMR methods applied to solids led to the development of pulse sequences whose effect of the spin system may bring about a situation corresponding to time inversion with a definite accuracy which is specified by the experimental conditions.

Schneider and Schmiedel (1969) were the first to notice the possibility of observation, using pulsed NMR methods, of the evolution of free induction of the system under conditions of a change in the time sign. The experiment suggested by Schneider and Schmiedel (1969) aimed at perfecting possibilities of measuring the second moment of the NMR line.

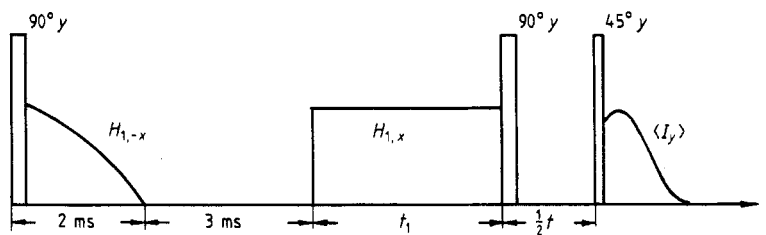


Figure 1. Pulse sequence used.

Waugh and co-workers (Rhim *et al* 1970, 1971) observed, on the basis of the pulse sequence suggested by Schneider and Schmiedel (1969), restoration of the free induction signal owing to the change in sign of the secular part of dipole–dipole interactions after a time lapse many times longer than the spin–spin relaxation time. The phenomenon observed by Rhim *et al* (1970, 1971) became known as ‘magic echo’.

Skrebnev and Safin (1986) and Skrebnev *et al* (1987) developed pulse sequences that made it possible to observe restoration of the dipole signal under conditions of a change in the time sign or the dipole magic echo. The distinguishing feature of this phenomenon compared with the magic echo of the free-induction signal is that in the case of the dipole magic echo when there is a change in the interaction sign in the Hamiltonian of the system, evolution can be observed in the direction opposite to the time of the term in the macrosystem density matrix which also owes its origin to interactions within this system.

With the help of the dipole magic echo Skrebnev and Safin (1986) compared the behaviour under conditions of time inversion of the operators in the density matrix to which the energy reservoir corresponds and of the operators in the energy representation having only non-diagonal matrix elements. The experiments demonstrated that the evolution of the non-diagonal operators is well described by the quantum-mechanical equations of motion. At the same time peculiarities in the behaviour of the operator to which the energy reservoir corresponds cannot be explained on the basis of reversible equations of mechanics. As pointed out by Skrebnev and Safin (1986) the approach in this case should be based on the non-equilibrium thermodynamics methods. It is the object of this paper to put forward this kind of approach.

2. Thermodynamical description of the irreversible component of the evolution of the dipole sub-system

To study the behaviour under conditions of time inversion of the energy reservoir of dipole–dipole interactions Skrebnev and Safin (1986) used the pulse sequence 1 shown in figure 1. Experiments were conducted on the nuclear spin system ^{19}F in the monocrystal CaF_2 at 300 K in an orientation [111] with respect to a constant magnetic field. Since the spin-lattice relaxation time of the sample under investigation was several seconds, the spin system of nuclei should be considered isolated from the lattice during the action of pulse sequence 1.

In the presence of a radio-frequency field of resonance frequency ω_0 in the tilted rotating frame, the transition into which is produced with the help of the operator

$\exp(-\frac{1}{2}i\pi I_y) \exp(i\omega_0 I_z t)$ the Hamiltonian of the spin system under study has the form (Skrebnev and Safin 1986)

$$\mathcal{H} = \omega_1 I_z - \frac{1}{2}\mathcal{H}'_d + \frac{3}{8}P, \tag{2}$$

where

$$\mathcal{H}'_d = \sum_{i<j} \mathcal{H}_{dij} = \sum_{i<j} a_{ij} [I_{zi} I_{zj} - \frac{1}{4}(I_{+i} I_{-j} + I_{-i} I_{+j})] \tag{3}$$

$$P = \sum_{i<j} P_{ij} = P^{(2)} + P^{(-2)} \tag{4}$$

$$P^{(2)} = \sum_{i<j} P_{ij}^{(2)} = \sum_{i<j} a_{ij} I_{+i} I_{+j} \quad P^{(-2)} = \sum_{i<j} P_{ij}^{(-2)} = \sum_{i<j} a_{ij} I_{-i} I_{-j}.$$

At the moment the radio-frequency field is imposed on the system, the density matrix in the tilted rotating frame will be written in the form

$$\sigma(0) = 1 - \beta(\frac{3}{8}P - \frac{1}{2}\mathcal{H}'_d). \tag{5}$$

From (1) it follows (Skrebnev and Safin 1986) that:

$$P(t_1 + t) = A_1^{-1} P A_1 \tag{6}$$

$$A_1 = \exp[i(\omega_1 I_z - \frac{1}{2}\mathcal{H}'_d + \frac{3}{8}P)t_1] \exp(i\mathcal{H}'_d t).$$

If $\omega_1 \gg \omega_L$ ($\omega_L = \gamma H_L$, where the local field $H_L = \{[\text{Tr}(\mathcal{H}'_d)^2]/[\text{Tr}(M_z^2)]\}^{1/2}$ (Goldman 1970) and $\gamma = 4005 \times 2\pi$) then in the case of $t = \frac{1}{2}t_1$ (6) is reduced to the form (Skrebnev and Safin 1986)

$$P(\frac{3}{2}t_1) = A_2^{-1} P A_2 \tag{7}$$

$$A_2 = \exp(i\omega_1 I_z t_1) T \exp\left(i \int_0^{t_1} \exp(-\frac{1}{2}i\mathcal{H}'_d t) \left(\frac{3}{8}\right)^2 \frac{[P^{(2)}, P^{(-2)}]}{2\omega_1} \exp(\frac{1}{2}i\mathcal{H}'_d t) dt\right)$$

where T is chronological operator.

In accord with (7) the amplitude of magic echo signal induced by the operator P ought to increase with the rise of ω_1 (Skrebnev and Safin 1986). Measurements showed, however, that contrary to the predictions of the theory based on equation (1) the amplitude of the signal in the pulse sequence 1 does not depend on ω_1 . To account for this fact one should take note that during the action of the radio-frequency field a similar operator in the Hamiltonian (2) corresponds to the operator $\frac{3}{8}P$ in the density matrix. In other words, a macroscopic energy reservoir whose evolution is to proceed in keeping with the second law of thermodynamics corresponds to the operator $\frac{3}{8}P$. To describe the behaviour of the subsystem $\frac{3}{8}P$ let us turn to non-equilibrium thermodynamics in which in order to obtain irreversible equations from reversible equations of mechanics probability assumptions are necessarily introduced in one form or another.

From the standpoint of thermodynamics, during the action of the radio-frequency field when the Hamiltonian has the form (2) the density matrix should irreversibly evolve toward the form

$$\sigma = 1 - \beta_{eq}(\omega_1 I_z - \frac{1}{2}\mathcal{H}'_d + \frac{3}{8}P). \tag{8}$$

Experiments show that during a short time (of the order of several spin-spin relaxation times T_2) heat mixing of the Zeeman reservoir $\omega_1 I_z$ and the non-secular spin-spin

interaction reservoir occurs (Goldman 1970, Safin *et al* 1984) which results in the density matrix assuming the form

$$\sigma = 1 - \beta_z(\omega_1 I_z + \frac{3}{2}P) + \beta_d \frac{1}{2} \mathcal{H}'_d. \tag{9}$$

The process of levelling of the inverse temperatures is β_z and β_d significantly slower (Goldman 1970, Safin *et al* 1984).

The evolution of the initial density matrix (5) will be described by means of the Liouville equation written in the form (Zubarev 1980)

$$\{\partial/\partial t + (1 - \mathcal{P}(t)) iL\} \Delta\sigma(t) = -(1 - \mathcal{P}(t)) iL \sigma_q(t). \tag{10}$$

Here L is the Liouville operator determined by the relationship

$$\begin{aligned} iLA &= -(1/i)[\mathcal{H}, A], \\ \Delta\sigma(t) &= \sigma(t) - \sigma_q(t), \\ \sigma_q(t) &= \exp\left(-\varphi(t) - \sum_n \mathcal{F}_n(t) P_n\right). \end{aligned} \tag{11}$$

We believe the set of operators P_n to be sufficient for a brief description of this non-equilibrrious state and the parameters $\mathcal{F}_n(t)$ are selected from the condition of equality of mean values of magnitudes P_n and their quasi-equilibrrious mean values

$$\langle P_n \rangle^t = \langle P_n \rangle^t_q = \text{Tr}(\sigma_q(t) P_n). \tag{12}$$

The effect of the Kawasaki–Guntton projection operator $\mathcal{P}(t)$ appearing in (10) on some operator A is determined as follows:

$$\begin{aligned} \mathcal{P}(t)A &= \sigma_q(t) \text{Tr} A + \sum_n (\text{Tr}(AP_n) - (\text{Tr} A)\langle P_n \rangle^t) \frac{\partial \sigma_q(t)}{\partial \langle P_n \rangle^t} \\ \frac{\partial \sigma_q(t)}{\partial \langle P_n \rangle^t} &= \sum_m \frac{\partial \sigma_q(t)}{\partial \mathcal{F}_m(t)} \frac{\partial \mathcal{F}_m(t)}{\partial \langle P_n \rangle^t}. \end{aligned} \tag{13}$$

It is natural in our case to choose as the set of P_n values the operators

$$\begin{aligned} P_1 &= \mathcal{H}_1 = \omega_1 I_z, \\ P_2 &= \mathcal{H}_2 = -\frac{1}{2} \mathcal{H}'_d, \\ P_3 &= \mathcal{H}_3 = \frac{3}{2} P. \end{aligned} \tag{14}$$

Then parameters \mathcal{F}_m having the meaning of inverse temperatures β_m of corresponding subsystems.

Since in our case $\sigma(0) = \sigma_q(0)$ the solution of the equation (10) has the form

$$\sigma(t) = \sigma_q(t) - \int_0^t T \exp\left(-\int_{t'}^t (1 - \mathcal{P}(t'')) iL dt''\right) (1 - \mathcal{P}(t')) iL \sigma_q(t') dt'. \tag{15}$$

In the case of high temperatures (undoubtedly achievable in our experiments)

$$\sigma_q(t) = \frac{1}{\text{Tr} \mathbf{I}} \left(1 - \sum_n \beta_n(t) \mathcal{H}_n\right). \tag{16}$$

The self-congruence condition (12) with regard for (16) enables us to write the system of equations for inverse temperatures β_n

$$\langle \mathcal{H}_k \rangle \frac{d\beta_k(t)}{dt} = -i \int_0^t \sum_l \beta_l(t') \left\langle [\mathcal{H}_l, \mathcal{H}_k] T \exp\left(-\int_{t'}^t (1 - \mathcal{P}(t'')) iL dt''\right) iL \mathcal{H}_l \right\rangle dt'. \tag{17}$$

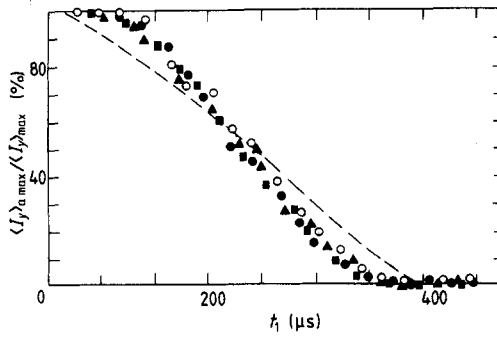


Figure 2. Dependence on t_1 of the signal maximum amplitude in the pulse sequence 1 at: $\omega_1/\gamma = 26\text{ G}$ (\circ), $\omega_1/\gamma = 19\text{ G}$ (\blacktriangle), $\omega_1/\gamma = 15\text{ G}$ (\bullet) and $\omega_1/\gamma = 13\text{ G}$ (\blacksquare). The broken curve corresponds to the solution of the equation (24) at $n = 0.25$ and $u^{1/2} = 7 \times 10^4\text{ s}^{-1}$.

When writing (17) we took into consideration that in our case $\mathcal{P}(t) iL \sigma_q(t) = 0$ and introduced the notation

$$\langle \dots \rangle = \frac{\text{Tr}(\dots)}{\text{Tr} \mathbf{1}}.$$

Immediate calculations yield

$$\begin{aligned} & \left\langle [\mathcal{H}_k, \mathcal{H}_k] T \exp\left(-\int_{t'}^t (1 - \mathcal{P}(t'')) iL dt''\right) iL \mathcal{H}_i \right\rangle \\ &= \langle [\mathcal{H}_k, \mathcal{H}_k] \exp(iL(t' - t)) iL \mathcal{H}_i \rangle \\ &+ \left\langle [\mathcal{H}_k, \mathcal{H}_k] i \sum_m \int_0^{t'-t} f_{ml}(t_1) \exp(iL(t' - t - t_1)) \mathcal{H}_k dt_1 \right\rangle \\ &= i \left\langle [\mathcal{H}_k, \mathcal{H}_k] \left([\mathcal{H}_k, \mathcal{H}_i](t' - t) + \sum_m \int_0^{t'-t} f_{ml}(t_1) \mathcal{H}_m(t' - t - t_1) dt_1 \right) \right\rangle \end{aligned} \tag{18}$$

where f_{ml} arise when summing up all \mathcal{H}_m factors obtained as a result of the expansion of the time-ordered exponent and projection with the help of the operator \mathcal{P} .

When recovering correlation functions (18) it should be taken into account that the possibility of describing the state of the subsystems \mathcal{H}_i by means of the temperatures β_i^{-1} arises owing to the decay during some time t_c of correlations between the states of various objects making up the subsystem or cross-correlations. In our case the time t_c is determined by the time of mutual reorientation of interacting spins i.e. is of the order of $20\ \mu\text{s}$. The decay of cross-correlations results in statistical independence of states of objects in the subsystem and ensures its maximum entropy. Physical admissibility of the assumption of the cross-correlations decay is valid in the case when the mean energy of heat motion exceeds the mean energy of interaction on a per-particle basis as in the conditions of our experiments. The cross-correlation decay refers, evidently, to energy

fluxes as well. Thus a consistent approach requires that the terms corresponding to cross-correlations be absent in correlation functions when using quasi-equilibrium operator (16). Accordingly we get

$$\langle \mathcal{H}_1^2 \rangle \frac{d\beta_1}{dt} = \int_0^t (\beta_3(t') - \beta_1(t')) G_{3,1}(t' - t) dt' + g_1(t) \quad (19)$$

$$\langle \mathcal{H}_2^2 \rangle \frac{d\beta_2}{dt} = \int_0^t (\beta_3(t') - \beta_2(t')) G_{3,2}(t' - t) dt' + g_2(t)$$

$$\langle \mathcal{H}_3^2 \rangle \frac{d\beta_3}{dt} = \int_0^t (\beta_1(t') - \beta_3(t')) G_{3,1}(t' - t) dt' + \int_0^t (\beta_2(t') - \beta_3(t')) G_{3,2}(t' - t) dt' + g_3(t).$$

Here

$$G_{3,1}(t' - t) = -4\left(\frac{3}{8}\right)^2 \omega_1^2 \sum_{i < j} \langle (P_{ij}^{(2)} - P_{ij}^{(-2)})(P_{ij}^{(2)} - P_{ij}^{(-2)})(t' - t) \rangle$$

$$G_{3,2}(t' - t) = -\frac{1}{4}\left(\frac{3}{8}\right)^2 \sum_{i,j,k,l} \langle [\mathcal{H}_{dij}, P_{kl}] [\mathcal{H}_{dij}, P_{kl}](t' - t) \rangle \quad (20)$$

$$g_k(t) = \int_0^t \sum_{l,m} dt' \beta_l(t') \int_0^{t'-t} dt_1 f_{ml}(t_1) \langle [\mathcal{H}_l, \mathcal{H}_k] \mathcal{H}_m(t' - t - t_1) \rangle_1.$$

In the correlation functions $\langle [\mathcal{H}_l, \mathcal{H}_k] \mathcal{H}_m(t' - t - t_1) \rangle_1$, whose form we do not write out in detail, cross-correlation terms are also absent.

Goldman (1970) pointed out that to an accuracy of the order of ω_L/ω_1 operators \mathcal{H}_Z and $\mathcal{H}_{n\text{-sec}}$ are related by a canonical transformation of the first order whose operator can be presented in our case in the form

$$R = i 3(P^{(2)} - P^{(-2)})/16\omega_1. \quad (21)$$

Transformation with the help of the operator R yields, with accuracy stated above:

$$\langle (P_{ij}^{(2)} - P_{ij}^{(-2)})(P_{ij}^{(2)} - P_{ij}^{(-2)})(t) \rangle$$

$$= \langle \{ P_{ij}^{(2)} - P_{ij}^{(-2)} + (i3/16\omega_1)[(P_{ij}^{(2)} - P_{ij}^{(-2)}), (P^{(2)} - P^{(-2)})] \}$$

$$\times \{ P_{ij}^{(2)} - P_{ij}^{(-2)} + (i3/16\omega_1)[(P_{ij}^{(2)} - P_{ij}^{(-2)}), (P^{(2)} - P^{(-2)})] \} (t^*) \rangle \quad (22)$$

$$\{ \dots \} (t^*) = \exp(-i\mathcal{H}^* t) \{ \dots \} \exp(i\mathcal{H}^* t)$$

$$\mathcal{H}^* = (1 - iR)\mathcal{H}(1 + iR) = \omega_1 I_z - \frac{1}{2}\mathcal{H}'_d + [(\frac{3}{8}P - \frac{1}{2}\mathcal{H}'_d), R].$$

Neglecting in \mathcal{H}^* terms of the order ω_1^2/ω_1 we write

$$\langle (P_{ij}^{(2)} - P_{ij}^{(-2)})(P_{ij}^{(2)} - P_{ij}^{(-2)})(t) \rangle = -e^{-2i\omega_1 t} \langle P_{ij}^{(2)} e^{i\mathcal{H}'_d t/2} P_{ij}^{(-2)} e^{-i\mathcal{H}'_d t/2} \rangle$$

$$- e^{2i\omega_1 t} \langle P_{ij}^{(-2)} e^{i\mathcal{H}'_d t/2} P_{ij}^{(2)} e^{-i\mathcal{H}'_d t/2} \rangle$$

$$- (2/\omega_1^2) (\frac{3}{16})^2 \langle [P_{ij}^{(2)}, P^{(-2)}] e^{i\mathcal{H}'_d t/2} [P_{ij}^{(-2)}, P^{(2)}] e^{-i\mathcal{H}'_d t/2} \rangle. \quad (23)$$

For $t > t_c$ integration over the time leads to averaging of oscillating terms to small values. The function $g_3(t)$ is also small, which is not difficult to see by using, when finding

correlation functions $\langle [\mathcal{H}, \mathcal{H}_k] \mathcal{H}_m(t) \rangle_1$, the transformation determined by the operator R (21), in a manner similar to that used in (22) and (23). As a result at $\omega_1 \gg \omega_L$ when $G_{3,1} \gg G_{3,2}$ and when β_1 may be put equal to zero, taking into account initial conditions (5), we find

$$\frac{d\beta_3}{dt} = - \int_0^t \beta_3(t') G_{3,1}^{(0)}(t' - t) dt' \quad (24)$$

where

$$G_{3,1}^{(0)}(t' - t) = \frac{3^2}{8^2 \langle P^{(2)} P^{(-2)} \rangle} \sum_{i < j} \langle [P_{ij}^{(2)}, P^{(-2)}] \times e^{i\mathcal{H}'_d(t'-t)/2} [P_{ij}^{(-2)}, P^{(2)}] e^{-i\mathcal{H}'_d(t'-t)/2} \rangle. \quad (25)$$

From (25) it can be seen that the rate of change of inverse temperature β_3 is independent of ω_1 . This corresponds to independence of ω_1 of the irreversible component of change in the system state derived when using pulse sequence 1.

It is not possible to compute the function $G_{3,1}^{(0)}$ accurately. However, let us put it in the form

$$G_{3,1}^{(0)}(t' - t) = (n\omega_1)^2 G(t' - t) \quad (26)$$

where n does not exceed unity by an order of magnitude.

Let us use for $G(t' - t)$ the Haussian approximation, which is natural for regular magnetics of the type CaF_2 (Abragam 1961, Goldman 1970)

$$G(t' - t) = \exp(-\frac{1}{2}M(t' - t)^2). \quad (27)$$

The value M is comparable in magnitude with the second moment of the NMR line in CaF_2 . In the orientation [111] for CaF_2 the square root of the second moment $M^{1/2} = 4 \times 10^4 \text{ s}^{-1}$ (Abragam 1961). Accordingly $\omega_L = (1/\sqrt{3})M^{1/2} = 2.35 \times 10^4 \text{ s}^{-1}$. The dotted line in figure 2 corresponds to the solution of the equation (24) at $n = 0.25$ and $M^{1/2} = 7 \times 10^4 \text{ s}^{-1}$. One can see that the equation (24), when the values of parameters correspond to the investigated sample, leads to a good agreement between the dependence on the time of the reversal temperature β_3 and the signal observed when employing pulse sequence 1.

If no account is taken of the cross-correlation decay then instead of $G_{3,1}(t)$ in the equations (19) there will be present correlation function

$$G_{3,1}^{(1)}(t) = -4(\frac{3}{8})^2 \omega_1^2 \langle (P^{(2)} - P^{(-2)})(P^{(2)} - P^{(-2)})(t) \rangle. \quad (28)$$

The transformation by means of the operator R (21) reduces (28) to the form

$$G_{3,1}^{(1)}(t) = -4(\frac{3}{8})^2 \omega_1^2 \langle (P^{(2)} - P^{(-2)})(P^{(2)} - P^{(-2)})(t^*) \rangle \\ = 4(\frac{3}{8})^2 \omega_1^2 \{ e^{2i\omega_1 t} \langle P^{(-2)} e^{i\mathcal{H}'_d t/2} P^{(2)} e^{-i\mathcal{H}'_d t/2} \rangle + e^{-2i\omega_1 t} \langle P^{(2)} e^{i\mathcal{H}'_d t/2} P^{(-2)} e^{-i\mathcal{H}'_d t/2} \rangle \}. \quad (29)$$

It is obvious that the equations (19) with the correlation function $G_{3,1}^{(1)}$ (29) cannot describe the rapid and ω_1 -independent approach of the inverse temperature β_3 to the value β_1 .

† In the paper by Skrebnev and Safin (1986), figure 4 shows $\langle I_y \rangle_{a\text{max}} / \langle I_y \rangle_{\text{max}}$ falling to 50%. This discrepancy in the notation, which is equivalent to taking into consideration the non-essential constant contribution to the signal, is corrected in the present paper, where figure 2 shows $\langle I_y \rangle_{a\text{max}} / \langle I_y \rangle_{\text{max}}$ falling to zero.

In the book by Goldman (1970) the heat mixing process of the Zeeman and non-secular subsystems \mathcal{H}_Z and $\mathcal{H}_{n\text{-sec}}$ is treated on the basis of the expression (1). The contribution of the density matrix term $\beta\mathcal{H}_{n\text{-sec}}$ to magnetisation $\langle I_z \rangle$ is computed

$$\langle I_z \rangle = -\beta \text{Tr}\{I_z \exp(-iHt)\mathcal{H}_{n\text{-sec}} \exp(i\mathcal{H}t)\}. \quad (30)$$

However, it is impossible to describe the experimental situation resulting from the use of pulse sequence 1 or to derive the irreversible component of the non-secular subsystem evolution by means of (30).

3. Conclusion

The description of the behaviour of sub-system \mathbb{P} under conditions of time inversion proved possible when going from expression (1) to the methods of non-equilibrium thermodynamics. The probability assumptions, which allow us to take into consideration irreversibility in the evolution of the system and which lead to agreement with the experiment, were introduced by way of describing the state of sub-systems with the help of temperatures and taking account of autocorrelation only when finding correlation functions.

The necessity of involving probability assumptions to explain the results of the time inversion experiments follows from the existence of the source of irreversibility of a microscopic nature. Questions about the nature of this source, the ways it is reflected in the equations of motion and its role in systems with a small number of particles remain open.

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